PB88-190483

Review of Methods for Remote Sensing of Atmospheric Emissions from Stationary Sources

Research Triangle Inst.
Research Triangle Park, NC

Prepared for

Environmental Monitoring Systems Lab.
Research Triangle Park, NC

Mar 88

U.S. Department of Commerce
National Technical Information Service
NTIS

TECHNICAL REPORT DATA

(Please read Instructions on the reverse before completing)

1. REPORT NO.
EPA/600/4-88/014

2.

3. RECIPIENT'S ACCESSION NO.
PB88 190483/AS

4. TITLE AND SUBTITLE
A Review of Methods for Remote Sensing of Atmospheric Emissions from Stationary Sources

5. REPORT DATE
March 1988

6. PERFORMING ORGANIZATION CODE

7. AUTHOR(S)
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8. PERFORMING ORGANIZATION REPORT NO.

9. PERFORMING ORGANIZATION NAME AND ADDRESS
* Research Triangle Institute
Research Triangle Park, NC 27709

** Southern Research Institute
Birmingham, AL 55305

10. PROGRAM ELEMENT NO.

11. CONTRACT/GRANT NO.
68-02-4442

12. SPONSORING AGENCY NAME AND ADDRESS
U.S. Environmental Protection Agency
Office of Research and Development
Environmental Monitoring Systems Laboratory
Research Triangle Park, NC 27711

13. TYPE OF REPORT AND PERIOD COVERED
Final

14. SPONSORING AGENCY CODE
EPA-600/08

15. SUPPLEMENTARY NOTES

16. ABSTRACT

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This report reviews the commercially available and developing technologies for the application of remote sensing to the measurement of source emissions. Many remote monitoring techniques are applicable to source measurements. In general, passive techniques are easier to use and less expensive to purchase and operate than are the active systems. Active systems are, in general, more specific and more sensitive than the passive systems. Some active techniques provide the added advantage of range resolution, which is preferred for some types of measurement applications. The results of the study suggest that the U.S. Environmental Protection Agency should consider further study of remote sensing technology for some applications related to source monitoring.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

18. DISTRIBUTION STATEMENT
Release to public

19. SECURITY CLASS *(This Report)*
Unclassified

20. SECURITY CLASS *(This page)*
Unclassified

21. NO. OF PAGES
98

22. PRICE

EPA Form 2220-1 (Rev. 4-77) PREVIOUS EDITION IS OBSOLETE

PB88-190483

RTI/3744/04-01F

EPA/600/4-88/014
March 1988

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EPA Project Number 68-02-4442
Work Assignment 4

Project Officer

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OFFICE OF RESEARCH AND DEVELOPMENT
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U.S. DEPARTMENT OF COMMERCE
National Technical Information Service
SPRINGFIELD, VA. 22161

DISCLAIMER

The information in this document has been funded wholly by the U.S. Environmental Protection Agency under Contract 68-02-4442 to Research Triangle Institute. It has been subjected to the Agency's peer and administrative review, and it has been approved for publication as an Agency document. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

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CONTENTS

Section | Page

Abstract.......... iii
List of Figures.......... v
List of Tables.......... v
Abbreviations and Symbols vi

1. Introduction.......... 1

2. Techniques and Principles.......... 3
    Techniques for Remote Sensing.......... 3
    Principles of Remote Sensing.......... 26

3. Summary of Remote Sensing Technologies.......... 30
    Commercially Available Instruments.......... 30
    Developing Technologies.......... 46
    Potential Developments.......... 58

4. Practical Considerations.......... 60
    General.......... 60
    Advantages and Limitations.......... 61

References.......... 63

Appendix.......... 73

FIGURES

Number		Page
1	Diagram of a simplified Michelson interferometer and a graph of the output light intensity as a function of the displacement of the moving mirror........................	8
2	Photometer trace for NO_2 absorption and mask for measurement of NO_2 in a mask correlation spectrometer........	11
3	Nondispersive trace-gas analyzer.............................	13
4	Basic DIAL lidar system configuration........................	19
5	Example of wedge absorption data representation..............	25
6	Components of radiation involved in passive remote measurement of emission..	28
7	Correct cursor pattern position for plume measurement with a radiometer...	31
8	Sulfur dioxide comparative measurements by visiplume and reference methods..	32
9	Linear regression analysis for each major test type..........	34
10	ROM-I versus transmissometer data for a smoke generator plume...	35
11	EPA ROSE infrared spectrometer system........................	40

TABLES

1 | Threshold Limit Values for Direct Ocular Exposures (Intrabeam Viewing) from a Laser Beam........................ | 5
2 | Current BAGI CO_2 Laser Gas Absorption Data Base and Detection Sensitivity Estimates.............................. | 59

ABBREVIATIONS AND SYMBOLS

A	ampere
Å	angstrom
ac	alternating current
ACGIH	American Conference of Governmental Industrial Hygienists
ANSI	American National Standards Institute
BAGI	backscatter/absorption gas imaging
°C	degree Celsius
CEC	Commission of European Communities
CGC	Computer Genetics Corporation
cm	centimeter
DIAL	differential absorption lidar
DISC	differential scatter (lidar)
DOAS	differential optical absorption spectroscopy
ELS	Environmental Laser Systems, Incorporated
Er:YAG	erbium:ytterbium-aluminum-garnet
°F	degree Fahrenheit
FLIR	forward-looking infrared
FTIR	Fourier transform infrared
GASPILS	gas spill leak sensor
GRI	Gas Research Institute
h	hour
Hz	hertz
in.	inch
IRGIS	infrared gas imaging system
IRIS	Infrared Instrumentation Society
J	joule
kg	kilogram
km	kilometer
kW	kilowatt

kΩ	kilohm
LASERSAFE	laser remote sensing of atmospheric fugitive emissions
lidar	light detection and ranging
LOPAIR	long-path infrared
m	meter
μm	micrometer
μs	microsecond
μW	microwatt
mA	milliamperes
mg/m^3	milligram per square meter
mJ	millijoule
mm	millimeter
mW	milliwatt
min	minute
mrad	milliradian
NASA	National Aeronautics and Space Administration
NASA/Goddard	Goddard Space Flight Center
NASA/Langley	Langley Research Center
Nd:YAG	neodymium-doped yttrium-aluminum-garnet
nm	nanometer
NO_2	nitrogen dioxide
ns	nanosecond
ppb	parts per billion
ppm	parts per million
ppm-m	parts per million meter
ppt	parts per trillion
pW	picowatt
ROSE	remote optical sensing of emissions
s	second
SO_2	sulfur dioxide
SRI	SRI International, Inc.
TDL	tunable diode laser
TLV	threshold limit value
TWA	time-weighted average

U.S. EPA	United States Environmental Protection Agency
V ac	volts, alternating current
V dc	volts, direct current
W	watt
Ω	ohm

SECTION 1

INTRODUCTION

The concept of measuring atmospheric trace species by remote sensing has been actively pursued for the past 20 years. Remote sensing of the atmosphere involves determining the concentration of trace species, or optical properties caused by the presence of a species, in a region of the atmosphere that is removed from the detection equipment. Applications of remote sensing range from satellite-mounted systems[1] to portable, passive optical receivers.[2]

The term "remote sensing," as applied in this report, implies the detection or concentration measurement of trace atmospheric species by sensing the interaction of propagating electromagnetic energy and the specific constituent along the path of propagation. The source of the energy can be natural or designed as part of the system. In this report, those techniques that rely on a natural radiation source are considered passive techniques, and those that include their own radiation source are referred to as active techniques.

The purpose of this report is to describe the operational characteristics and applications of passive and active remote sensing technologies that are effective as ground-based source emission monitors. The review includes detection capabilities (e.g., range, sensitivity, accuracy, interferences), operational concerns (e.g., size, weight, mobility, power requirements), and, whenever possible, the capital and operating costs of the systems. Section 2 of the report discusses the operational principles of the various remote sensing techniques in a generic sense, as well as other background information related to remote sensing. Section 3 describes the available remote sensing systems and includes subsections on commercially available technology, developing techniques in the testing and demonstration stage, and anticipated emerging technologies. Section 4 summarizes the findings.

This report does not describe every remote measurement system that has been used for or is capable of measuring air pollution emissions. It does, however, describe the primary technologies used in remote atmospheric monitoring and gives examples of techniques that are applied to air pollution emission monitoring. Some innovative applications that will likely extend the capabilities of remote sensing to more uses are also described.

It should be noted that an earlier U.S. EPA report on this subject was published in 1976.[3] The interested reader should consult that report for additional information concerning the application of remote techniques in stationary source air emission monitoring. This report contains information that was not available in 1976.

In recent years, several review articles have addressed the more general topic of the application of remote sensing techniques to atmospheric monitoring. Grant and Menzies presented a survey of remote sensing of pollutant gases based on the differential absorption light detection and ranging (lidar) technique, or DIAL.[4] They divided the discussion of active laser remote sensors by the type of laser that is used to provide the excitation light. Within each division, the specific pollutants that are measurable were mentioned. Emphasis was placed on the best field measurement systems and results to data, and on promising laboratory developments. Finally, several passive optical remote sensors were discussed briefly. Killinger and Menyuk covered essentially the same ground in their review article.[5] However, their discussion emphasized the general types of optical interaction and the general lidar classes that are available for remote sensing rather than the specific laser types. They also included a section on the accuracy limitations of lidar measurements. Both articles provide useful information about remote sensing methods.

Although not specifically a discussion of remote sensing methods, Grigg and Ludwig's article on the legal and technical aspects involved in the application of remote sensing in air enforcement programs should also be noted.[6] In general terms, they discuss the technical characteristics that a remote sensor must have for its data to be admissible evidence in a court of law. This aspect of remote sensing has been ignored in most articles on the topic.

SECTION 2

TECHNIQUES AND PRINCIPLES

TECHNIQUES FOR REMOTE SENSING

The surveillance of air pollutant source emissions is of concern to regulators, process operators, process engineers, air pollution control engineers, and air pollution researchers. The primary objective of this research was to provide a summary of remote monitoring capabilities that have been used for, or are applicable to, air pollutant emissions monitoring. Many of these methods were identified.

The remote monitoring technologies reviewed in this report are limited to those that rely on the interaction of a light source and some atmospheric property to detect the presence of an atmospheric constituent. Remote monitoring technologies have been divided into passive systems and active systems. Passive systems use some natural light source; in active systems, a light source is included as a part of the instrument design.

Passive techniques for remote sensing, by definition, operate without the advantages of a controlled light source. They are designed to use naturally occurring light. The radiation upon which the analyses are based can be skylight, scenery light, direct sunlight, or thermally emitted light, depending on what is to be detected and the characteristics of the surroundings. These characteristics impose certain limitations on the monitoring capabilities. For example, all except those using thermally emitted radiation must be used during daylight periods, and some are further restricted by weather conditions. However, the advantage of simplicity, which passive sensors offer over active sensors, makes them a viable option for remote environmental monitoring. This is particularly true when conditions would favor the use of a portable, mobile instrument.

Passive sensors exist that operate in either or both of the two primary modes of molecular spectroscopy--emission and absorption. In the emission mode, radiation that is naturally emitted from the species target is directed to a detector and the signal is processed electronically. In the absorption mode, the sensors measure the transmittance characteristics of the emissions (or a related parameter) for sunlight or other natural light sources as the light passes through the atmosphere.

In the most general sense, active remote measurement techniques are those that rely on the interaction of the target atmospheric constituent with a controlled light source. The most common radiation sources used in active remote sensing are lasers. Lasers operate in a range of wavelengths spanning the infrared through ultraviolet regions, depending on the characteristics of the lasing medium. Some individual lasers can be tuned and can therefore operate over a range of wavelengths. By selecting a wavelength to coincide with an absorption or scattering feature of a constituent of interest, the laser beam can be made to interact with that constituent. The measurement of the magnitude of that interaction can be used to infer the concentration of the species along the path of the laser beam propagation. Other light sources can be used; however, lasers are, by far, the most popular light source because of their specific properties. Those properties include narrow spectral width (<0.01 nm), a frequency that is often tunable, and high pulse energy (up to 10 J per pulse for ruby lasers[5]) that can be pulsed rapidly (<1 μs) and focused into a narrow beam (<10 cm diameter).

Because of the power that can be produced by lasers, care must be taken to avoid eye damage caused by accidental exposure to the laser beam. The American National Standards Institute (ANSI) has established an acceptable-exposure-level standard for lasers,[7] and the American Conference of Governmental Industrial Hygienists (ACGIH) has established threshold limit values (TLVs) for laser exposure.[8,9] See also Reference 3 for a discussion of eye safety hazards from laser systems.

The TLVs are for exposure to laser radiation under conditions to which nearly all workers may be exposed without adverse effects. The values should be used as guides in the control of exposures and should not be regarded as fine lines between safe and dangerous levels. They are based on the best available information from exposure studies. Table 1 lists the TLVs for direct ocular exposures (intrabeam viewing) from a laser beam. The ACGIH also

TABLE 1. THRESHOLD LIMIT VALUES FOR DIRECT OCULAR EXPOSURES (INTRABEAM VIEWING) FROM A LASER BEAM

Spectral region[a]	Wavelength (nm)	Exposure time, t (s)	TLV (J/cm^2)
UVC	200 to 280	10^{-9} to 3×10^4	3.0×10^{-3} (b)
UVB	280 to 302	10^{-9} to 3×10^4	3.0×10^{-3} (b)
	303	10^{-9} to 3×10^4	4.0×10^{-3} (b)
	304	10^{-9} to 3×10^4	6.0×10^{-3} (b)
	305	10^{-9} to 3×10^4	1.0×10^{-2} (b)
	306	10^{-9} to 3×10^4	1.6×10^{-2} (b)
	307	10^{-9} to 3×10^4	2.5×10^{-2} (b)
	308	10^{-9} to 3×10^4	4.0×10^{-2} (b)
	309	10^{-9} to 3×10^4	6.3×10^{-2} (b)
	310	10^{-9} to 3×10^4	1.0×10^{-1} (b)
	311	10^{-9} to 3×10^4	1.6×10^{-1} (b)
	312	10^{-9} to 3×10^4	2.5×10^{-1} (b)
	313	10^{-9} to 3×10^4	4.0×10^{-1} (b)
	314	10^{-9} to 3×10^4	6.3×10^{-1} (b)
UVA	315 to 400	10^{-9} to 10^0	$5.6 \times 10^{-1}\ t^{1/4}$
	315 to 400	10^0 to 10^3	1.0
	315 to 400	10^3 to 3×10^4	1.0 (mW/cm^2)
Visible	400 to 700	10^{-9} to 1.8×10^{-5}	5.0×10^{-7}
	400 to 700	1.8×10^{-5} to 10^0	$1.8 \times 10^{-3}\ t^{3/4}$
	400 to 549	10^0 to 10^4	1.0×10^{-2}
	550 to 700	10^0 to T_1 (c)	$1.8 \times 10^{-3}\ t^{3/4}$
	550 to 700	T_1 to 10^4	$1.0 \times 10^{-2}\ C_B$ (d)
	400 to 700	10^4 to 3×10^4	C_B ($\mu W/cm^2$)
IRA	700 to 1,049	10^{-9} to 1.8×10^{-5}	$5.0 \times 10^{-7}\ C_A$ (e)
	700 to 1,049	1.8×10^{-5} to 10^3	$1.8 \times 10^{-3}\ C_A$
	1,050 to 1,400	10^{-9} to 10^{-4}	5.0×10^{-6}
	1,050 to 1,400	10^{-4} to 10^3	$9.0 \times 10^{-3}\ t^{3/4}$
	700 to 1,400	10^3 to 3×10^4	$3.2 \times 10^2\ C_A$ (W/cm^2)
IRB & IRC	1,400 to 10^6	10^{-9} to 10^{-7}	1.0×10^{-2}
	1,400 to 10^6	10^{-7} to 10^0	$5.6 \times 10^{-1}\ t^{1/4}$
	1,400 to 10^6	10^0 to 3×10^4	1.0×10^{-1} (W/cm^2)

[a] Regions of the optical radiation spectrum are designated as follows:

Region	Wavelength range
Ultraviolet (UV)	100 to 380-400 nm
UVC	100 to 280 nm
UVB	280 to 315-320 nm
UVA	315-320 to 380-400 nm
Light (visible)	380-400 to 760-780 nm
Infrared (IR)	760-780 nm to 1 mm
IRA	760-780 nm to 1.4 μm
IRB	1.4 to 3.0 μm
IRC	3.0 μm to 1 mm

[b] Not to exceed $5.6 \times 10^{-1}\ t^{1/4}\ J/cm^2$ for $t \leq 10$ s.

[c] T_1 = 10 s for wavelengths (λ) from 400 to 549 nm;
= $10 \times 10^{[0.02(\lambda-550)]}$ for λ from 550 to 700 nm.

[d] C_B = 1 for λ from 400 to 549 nm;
= $10^{[0.015(\lambda-550)]}$ for λ from 550 to 700 nm.

[e] $C_A = 10^{[0.002(\lambda-700)]}$ for λ from 700 to 1,049 nm;
= 5 for λ from 1,050 to 1,400 nm.

has established TLVs for viewing a diffuse reflection of a laser beam or an extended-source laser and for skin exposure from a laser beam. There is also a TLV for exposure from repetitively pulsed lasers. The TLVs may be averaged over an aperture of 1 mm, except for (1) TLVs for the eye in the spectral range from 400 to 1,400 nm, which should be averaged over a 7-mm limiting aperture (e.g., the pupil) and (2) all TLVs for the spectral region from 10^5 to 10^6 nm, which should be averaged over a 10-mm limiting aperture. No modification of the TLVs is permitted for pupil sizes less than 7 nm.

Grant and Hake demonstrated the feasibility of using the DIAL technique for range-resolved measurement of sulfur dioxide and ozone at ultraviolet wavelengths with an eye-safe lidar.[10] Their power levels were low enough that a modest beam expander was sufficient to make their lidar eye-safe at the transmitter output. A 0.75-mJ pulse at a wavelength of 292 nm used a 2-in. diameter transmitter to meet the ANSI standard.

An alternate approach is to use lasers that operate in the "eye-safe" spectral region of wavelengths greater than 1.4 μm. Carbon monoxide, carbon dioxide, and tunable diode lasers and optical parametric oscillators operate in this spectral region.

Passive Techniques

Radiometers--

Radiometers are nondispersive instruments that isolate the spectral range of interest through use of simple systems such as interference filters. Traditionally, they have been used in the ultraviolet and visible region[11] for situations requiring specificity. Spectral resolution is generally too poor for use in the infrared region except for qualitative information or when a particularly strong infrared absorber is known to be the only component that can produce a substantial signal. Applications of this technique for measuring emissions have recently been developed. These are discussed in Section 3. A correlation technique that greatly improves the spectral specificity of radiometers has also been developed and is discussed later in this section. Advancements in video technology have led to application of imaging systems to situations needing quantitative monitoring in the ultraviolet and infrared regions.

Spectrometers--

Spectrometers are wavelength dispersive instruments that use either a prism or a grating to separate the incoming radiation into its spectrum. Spectrometers, like radiometers, operate most successfully in the visible and ultraviolet regions. Insufficient resolution of spectrometers in the infrared region, compared to interferometers, has essentially eliminated conventional spectrometers from use in remote sensing applications.[12]

Interferometers--

Three types of interferometers have been designed for remote sensing applications: Fabry-Perot interferometers, Michelson interferometers, and Hadamard transform spectrometers. The Fabry-Perot interferometer, though technically a dispersive device, increases the throughput of incoming radiation by a factor of about 100.[13,14] This interferometer uses circular apertures instead of slits, as in grating instruments. It is capable of high resolution in the ultraviolet and visible regions.

The application of the Michelson interferometer (Figure 1) to remote sensing has greatly improved detection capabilities in the infrared region. This region is of particular value in monitoring for organic species because they tend to have several absorption bands in the infrared region. The Michelson interferometer consists of two perpendicular plane mirrors, one of which is fixed in place and one that can move along an axis perpendicular to its plane. A beam splitter is located at an angle of 45 degrees to both mirrors. The incoming radiation is split into two paths and then recombined after a path difference has been introduced. The position of the movable mirror determines whether the recombined beams will interfere constructively or destructively. As the mirror is moved, an interferogram is produced. Light entering the interferometer can be considered to be information encoded in a light-density versus wavelength domain. In the interferogram, this same spectral information is encoded in a light-density versus time domain. From this interferogram, a Fourier transformation is performed by computer to transform the time domain data to the wavelength domain. The Michelson interferometer provides a significant advantage in resolution in the IR region and in throughput in all spectral regions.[13]

Hadamard transform spectrometers have been built for ground-based automobile exhaust measurements and airborne applications.[16,17] Although a Hadamard

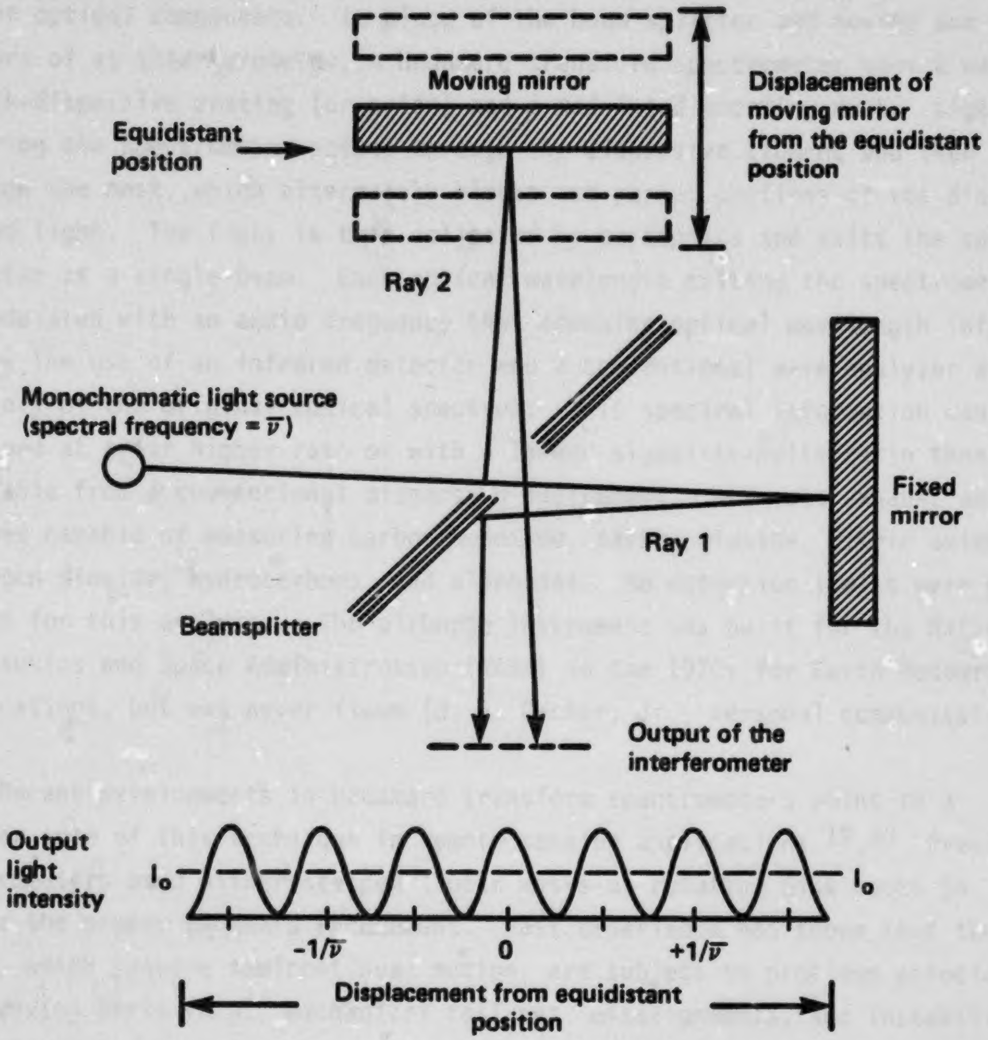
Moving mirror
Equidistant position
Displacement of moving mirror from the equidistant position
Ray 2
Monochromatic light source (spectral frequency = $\bar{\nu}$)
Fixed mirror
Ray 1
Beamsplitter
Output of the interferometer
Output light intensity
I_o
I_o
$-1/\bar{\nu}$
0
$+1/\bar{\nu}$
Displacement from equidistant position

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Figure 1. Diagram of a simplified Michelson interferometer and a graph of the output light intensity as a function of the displacement of the moving mirror.[15]

transform spectrometer is a multiplexing spectrometer, it is not an interferometer.[18] It is based on an entirely different mathematical concept to calculate the spectral distribution of incoming light. It also uses entirely different optical components. In place of the beam splitter and moving and fixed mirrors of an interferometer, a Hadamard transform spectrometer uses a wavelength-dispersive grating (or prism) and a modulated encoding mask. Light entering the spectrometer passes through the dispersive element and then through the mask, which alternately blocks and passes portions of the dispersed light. The light is then collected by postoptics and exits the spectrometer as a single beam. Each optical wavelength exiting the spectrometer is modulated with an audio frequency that contains optical wavelength information. The use of an infrared detector and a conventional wave analyzer allows recovery of the original optical spectrum. This spectral information can be obtained at a far higher rate or with a larger signal-to-noise ratio than is available from a conventional dispersing instrument. The auto exhaust analyzer was capable of measuring carbon monoxide, carbon dioxide, nitric oxide, nitrogen dioxide, hydrocarbons, and aldehydes. No detection limits were published for this analyzer. The airborne instrument was built for the National Aeronautics and Space Administration (NASA) in the 1970s for Earth Resources Applications, but was never flown (J. A. Decker, Jr., personal communication, 1987).

Recent developments in Hadamard transform spectrometers point to a greater role of this technique in remote sensing applications.[19,20] Previous spectrometers used either stepped linear masks or rotating disk masks to attain the proper Hadamard encodement. Past experience has shown that these masks, which require semicontinual motion, are subject to problems associated with moving parts (e.g., mechanical failures, misalignments, and instability in field environments). This was an unfortunate disadvantage, because the other components of the spectrometer not only were optically and mechanically rigid, but also were easily constructed with lower optical and mechanical tolerances than those for interferometers. The moving mask, therefore, tended to hamper the applicability of this technique to spectral analysis. The practical solution was to replace the moving mask with a stationary or solid-state mask fabricated from an electro-optic or thermodiachromatic material. The resulting spectrometer combined some of the better features of the single-detector, single-resolution-element spectrometers with some of the better features of the multiplexing spectrometers, while eliminating some of the disadvantages found in each spectrometric method. Hadamard transform spectrometers are currently being developed for application to volatile organic compound measurements at stationary sources (W. G. Fately, Kansas State University, personal communication, 1988).

Dispersive Correlation Techniques--

Correlation techniques are very useful in remote sensors because the hardware produces a direct, real-time measurement of the degree of correlation between the spectrum of the radiation coming from the target emissions and the known spectrum of a species to be detected. Mask correlation spectroscopy was begun in astronomy,[21,22] and its development for remote sensing has been extensive since then. In this technique, a mask is manufactured that resembles the spectrum of the species to be detected, and the incoming radiation from the target emissions is dispersed so that its spectrum is superimposed upon the mask at the entrance to the detector. In the simplest versions, the pattern on the mask, as illustrated in Figure 2, is a series of slits that are spaced corresponding to the wavelength spacing between absorption minima of the gas species to be detected.[23] Either the spectrum of the incoming radiation or the mask pattern is caused to oscillate very regularly and with constant amplitude so that the portion of the incoming spectrum that matches the spectrum of the gas to be detected causes the detector signal to oscillate at the frequency of the forced oscillation. The portion of the incoming radiation that does not match this spectrum appears as low-amplitude variations in the detector signal at lower and higher frequencies. The concentration of the species to be detected turns out to be proportional to the difference between the maximum and minimum of the detector signal referenced to its average.

Mask correlation spectrometers collect radiation from a suitable background, disperse it into its spectrum, and transmit it through masks before it reaches the detector. Skylight is usually employed as the background, but in some early prototypes, deuterium, quartz iodine, or xenon lamps were used as sources and measurements over long paths require the use of a lamp.[24,25] Because the mask is specific to the spectrum of the gas to be detected, one instrument can be used to monitor only one species at a time.

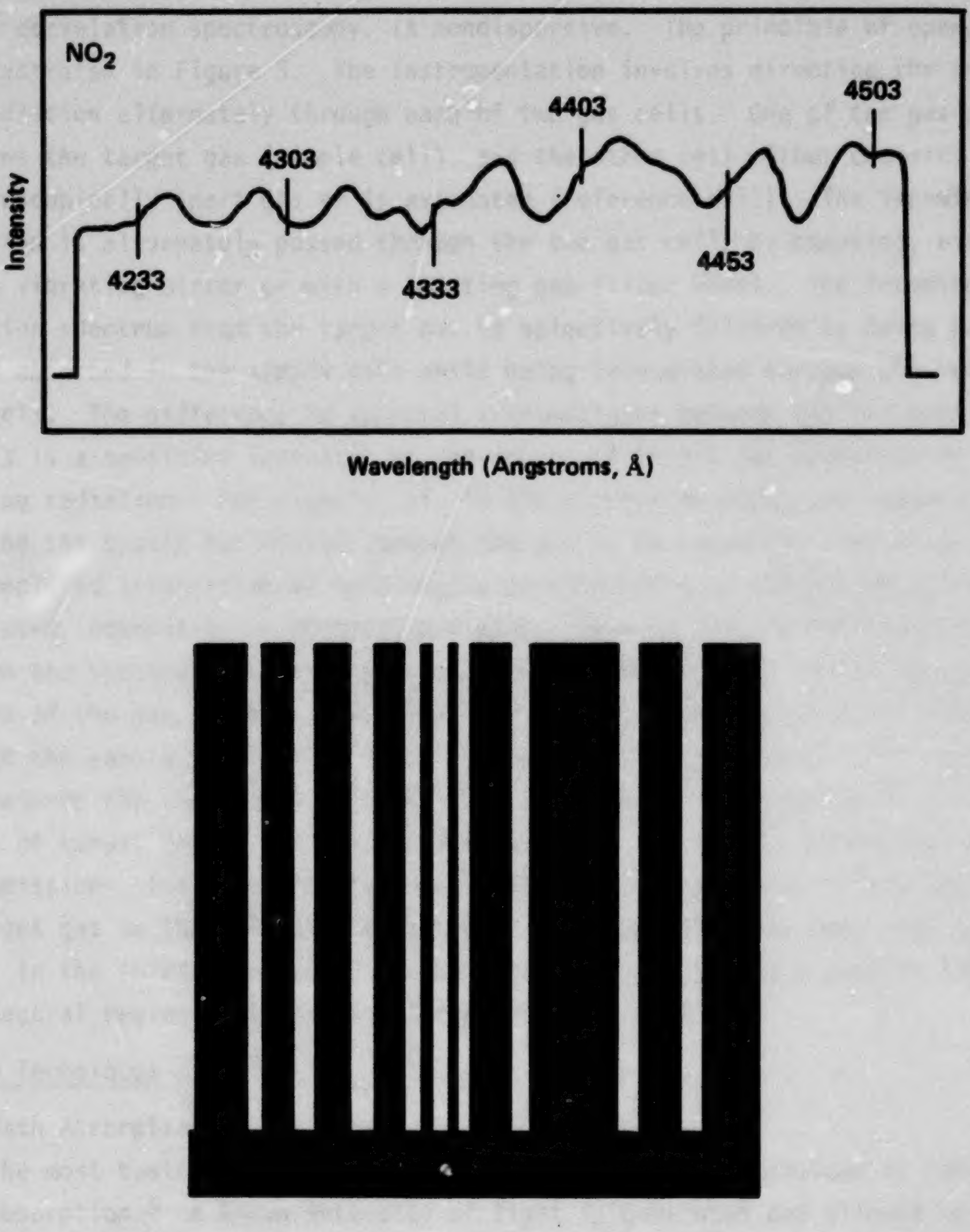

Figure 2. Photometer trace for NO_2 absorption and mask for measurement of NO_2 in a mask correlation spectrometer.[23]

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Nondispersive Correlation Techniques--

In a second type of correlation technique, a cell containing the target gas is used to mask the incoming radiation.[26,27] This technique, called gas-filter correlation spectroscopy, is nondispersive. The principle of operation is illustrated in Figure 3. The instrumentation involves directing the incoming radiation alternately through each of two gas cells. One of the gas cells contains the target gas (sample cell), and the other cell either contains a spectroscopically inert gas or is evacuated (reference cell). The incoming radiation is alternately passed through the two gas cells by chopping, either with a vibrating mirror or with a rotating gas-filter wheel. The incoming radiation spectrum from the target gas is selectively filtered by being partially absorbed in the sample cell while being transmitted through the reference cell. The difference in spectral transmittance between the two modulated signals is a sensitive indicator of the amount of target gas signature in the incoming radiation. For example, if, in the absorption mode, the radiation entering the system has passed through the gas to be detected, then it will have depleted intensities at wavelengths corresponding to absorption peaks and undepleted intensities at absorption minima. Because of this correlation between the spectral intensity of the incoming radiation and the absorption spectra of the gas in the sample cell, the radiant energy absorbed in passing through the sample gas cell is lower than would occur otherwise. The difference between the two modulated signals is then inversely proportional to the amount of target gas in the monitored emissions. In the mode employing radiation emissions, the signal difference is directly proportional to the amount of target gas in the monitored emissions. This technique has been used primarily in the infrared region. Spectral band-pass filters are used to isolate the spectral region of interest and improve sensitivity.

Active Techniques

Long-Path Absorption--

The most basic laser-based atmospheric measurements technique is long-path absorption.[5] A known intensity of light is generated and allowed to propagate through a predetermined space. The attenuation of light energy through the pathlength is assumed to be due to absorption by the target species. The measurement is an average burden over the pathlength and is commonly expressed

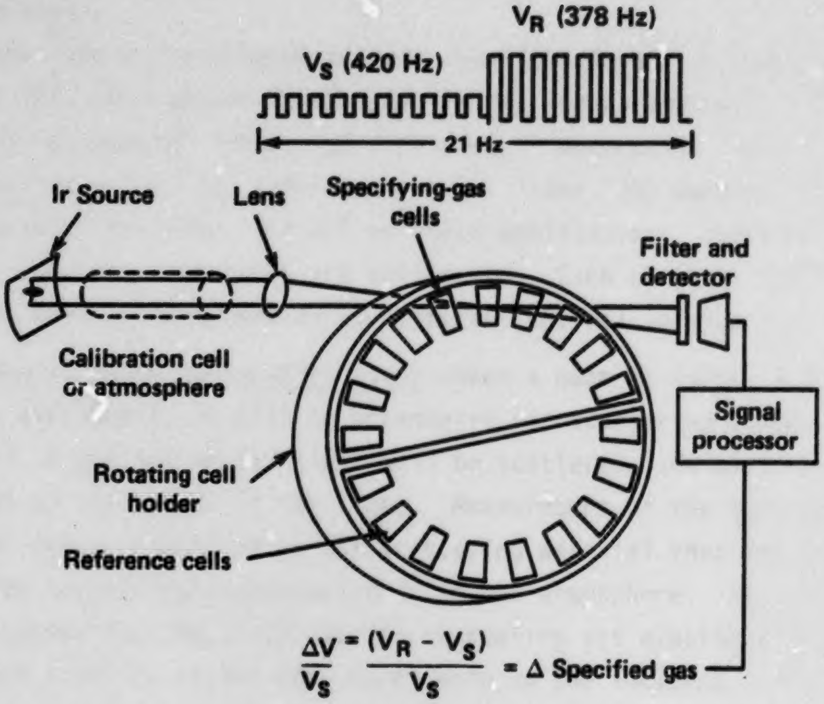

V_R (378 Hz)
V_S (420 Hz)
21 Hz
Ir Source
Lens
Specifying-gas cells
Filter and detector
Calibration cell or atmosphere
Signal processor
Rotating cell holder
Reference cells
ΔV/V_S = (V_R - V_S)/V_S = Δ Specified gas

Figure 3. Nondispersive trace-gas analyzer.[28]

as parts per million meters (ppm-m). The light source can be either positioned at a distance from the receiver and caused to shine across the sampling region to the receiver or positioned at the receiver and reflected by appropriately placed mirrors (or retroreflectors) back to the receiver. This particular technique can be applied with low-power light sources other than lasers.

Lidar Techniques--

The more common laser-based methods use light detection and ranging (lidar). Within this general technique are six common methods: (1) atmospheric (Mie) backscatter lidar, (2) differential absorption lidar (DIAL), (3) fluorescence lidar, (4) Raman backscatter lidar, (5) Doppler lidar, and (6) wedge absorption lidar. In all of these applications, the laser source and optical receiver components are collocated. Each of these lidar applications is designed to be sensitive to different optical interactions.

Atmospheric (Mie) backscatter lidar--When a beam of light is transmitted through the atmosphere, it will be attenuated via scattering processes and adsorption.[5] A portion of the light will be scattered back along the axis of illumination to the source of the light. Measurement of the backscattered light allows one to characterize the scattering material that the beam encountered. Three scattering processes occur in the atmosphere: Rayleigh, Mie, and Raman scattering. Rayleigh and Mie scattering are elastic processes and the scattered light is at the same wavelength as the incident light. Raman scattering is an inelastic process and the wavelength of the scattered light is red-shifted relative to that of the incident light. Raman scattering will be discussed further in a following section. Rayleigh and Raman scattering are associated with scattering material whose size is much smaller than the wavelength of the incident light (e.g., molecules in the atmosphere). When the scattering material is approximately the same size as the wavelength of the incident light (e.g., aerosols and the fine dusts in the atmosphere), Mie scattering occurs. For the visible and infrared spectral regions, the backscattered light is almost exclusively due to Mie scattering from particles.

Lidars have been constructed that use Mie backscattering to measure aerosol concentrations in the atmosphere and particulate emissions from stationary sources. Because Mie scattering is not strongly dependent on wavelength, untuned, pulsed lasers can be used as the light source. Ruby lasers with a wavelength of 694 nm and neodymium-doped yttrium-aluminum-garnet (Nd:YAG) lasers with a wavelength of 1,064 nm are commonly used for atmospheric backscatter lidars.

A significant portion of the research on the application of atmospheric backscatter lidar to stationary source measurements has been performed by Uthe and coworkers at SRI International, Inc. (SRI). Between 1971 and 1980, this group published a number of articles describing lidar observations of power plant plumes in Pennsylvania, Missouri, and Tennessee.[29-31] Although their instrumentation has evolved over the years, its basic design has remained the same. The entire system is operated from a truck or van provided with power-generating facilities to allow completely mobile operation or operations in remote locations. A Ruby or Nd:YAG laser is mounted such that elevation and azimuth can be varied. The backscattered light is gathered by a telescope and is detected by a photomultiplier tube. The data are recorded using video recording devices and subsequently are processed by computer. The lidar system is capable of plume measurements to distances of approximately 15 km and altitudes of approximately 3 km. It is able to track a plume to distances of up to 130 km from the stationary source. This system is also capable of calculating the mixing depth from measurements of atmospheric aerosol layers.[32] These calculations agree with those obtained from measurement by radiosondes and helicopter temperature soundings.

A similar atmospheric backscatter lidar was developed in the Federal Republic of Germany for remote measurements of plume dispersion over sea surfaces.[33] Although the components were generally the same as those of the SRI system, the lidar's transmitter and receiver weighed only 17 kg. The weight of the data-processing and recording components was not discussed. The lidar was capable of making measurements at distances from 1 to >2 km depending on the plume material.

An important application of atmospheric backscatter lidar has been the remote measurement of smoke plume transmittance. This van-mounted ruby lidar system was developed by the General Electric Company for the U.S. EPA.[34] It was developed for remote single-ended measurements of transmittance at ranges of 150 to 500 m. The measurement is obtained by comparing the clean-air lidar return from the near side of the plume with that from the far side. The lidar measurements showed good agreement with telephotometer measurements for three coal-fired power plants. They also showed good agreement with in-stack transmissometer measurements and with a trained opacity observer for nine coal-fired power plants, five oil-fired power plants, and several other stationary sources.[35]

Based on these and other studies, U.S. EPA has established atmospheric backscatter lidar as an alternate method for determining the opacity of emissions from stationary sources.[36-39] This fact is significant because atmospheric backscatter lidar is seen by U.S. EPA as an enforcement tool rather than a research tool. To date, U.S. EPA's National Enforcement Investigation Center has measured approximately 200 facilities using their Omega-1 lidar system, which was also fabricated by the General Electric Company (J. Hathaway, U.S. EPA, personal communication, 1988). These facilities include power plants, refineries, fertilizer plants, glass factories, and steel foundries. The center has participated in 30 enforcement actions in cooperation with U.S. EPA regional offices. These actions have resulted in 23 notices of violation being issued. The lidar system is currently in active use and is seen as a reliable working tool. Data from the lidar system is accepted in the courts as admissible evidence. This acceptance is seen to be a result of the quality assurance program that has been developed for the atmospheric backscatter lidar system.

Atmospheric backscatter lidar may also use multiple wavelengths of light to measure such parameters as smoke plume distribution, vertical transmission, and particle size. This approach is called differential scatter (DISC) lidar. Although Mie scattering does not depend strongly on wavelength, there is some dependence. The phenomenon can be used to investigate the backscattering arising from different-sized particles in the atmosphere. For example, near-infrared wavelengths (e.g., 1,065 nm) are very sensitive to low-density (subvisible) particulate pollution becasue of the larger ratio of aerosol-to-molecular scattering at longer wavelengths. Shorter wavelengths (e.g., 532 nm) are used to derive visibility-related information and to apply two-wavelength analysis of backscatter signals in terms of other aerosol parameters (e.g., extinction and concentration).

SRI International has developed several multiple-wavelength lidar systems.[40-42] A two-wavelength, downward-looking system was developed for airborne applications. This system was designed to observe smoke plume and boundary layer conditions over large regional areas. It has investigated forest fire plumes, the Los Angeles boundary layer, a power plant plume in complex terrain, and the effect of atmospheric convective processes on power plan plumes. The multiple-wavelength measurements yielded information on plume aerosol concentrations, mean particle size, and plume dispersion. SRI International conducted a feasibility study to investigate measuring sulfuric acid aerosols using this technique. A four-wavelength, van-mounted system has been used successfully for several unpublished applications for the U.S. Army Research Office (M. L. Wright, SRI, personal communication, 1987). An unpublished NASA feasibility study indicated that gaseous and particulate emissions from a single source can be measured by an orbiting sensor.

A similar two-wavelength lidar system has been developed in the Soviet Union.[43] It was used to measure aerosol mass concentrations approximately 2 km downwind of a metallurgical plant. Satisfactory agreement was obtained between the lidar measurements and data obtained from direct sampling of the plume from an aircraft.

Differential absorption lidar--DIAL is used for measuring gaseous atmospheric constituents. In this technique, two closely spaced laser wavelengths are monitored simultaneously. One wavelength is selected to coincide with an absorption feature of the target species. The second wavelength is just outside that absorption feature. The assumption of DIAL is that both wavelengths are equally affected by interferents, such as particles and water vapor, and therefore the difference in absorption between the two is due only to the effect of the target species.

It should be noted that one can obtain relatively large wavelength separations with some lasers. A group of the Kyushu University in Japan routinely uses a Raman-shifted laser pumped by a krypton fluoride excimer laser to generate light at 290, 313, and 360 nm.[44] This laser has been used for solar-blind DIAL measurements of ozone over an altitude range of 5 to 30 km. Such lasers may be applied to source-oriented DIAL measurements.

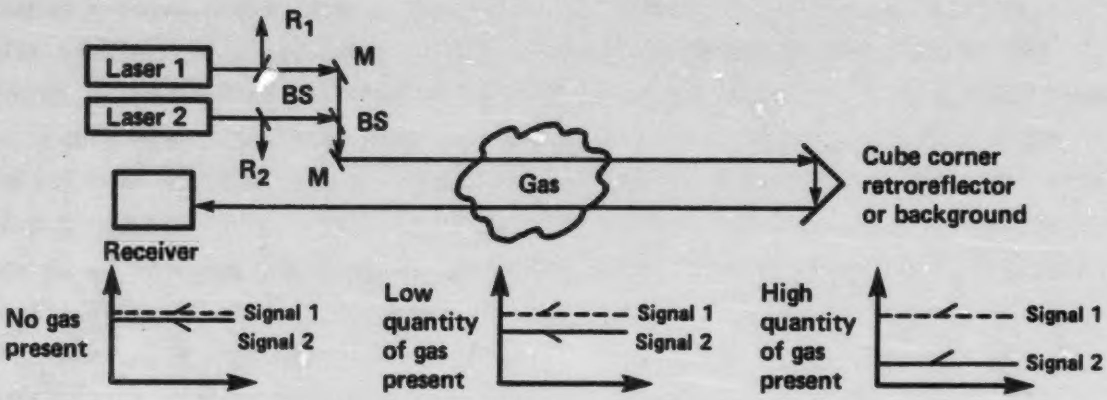
DIAL systems can be operated in two distinct modes that are distinguished from one another by the type of target that is used. The first mode is a long-path absorption system that uses a cooperative target (i.e., a retroreflector or topographic feature). In this mode, the measurement yields a mean concentration along the path between the DIAL system and the target. The second mode is a range-resolved system that uses the naturally occurring atmospheric aerosol as a distributed backscatter reflector. In this mode, the measurement yields concentrations as a function of the range from the DIAL system. The signal processing electronics are considerably more complex in this mode than in the first mode because the scattered signal returning to the DIAL system must be measured over a series of small time intervals.

The use of a retroreflector can lead to as much as an order of magnitude improvement in the concentration detection limit relative to the use of atmospheric backscatter. This improvement is due to the greater signal level that is returned by the retroreflector. It should be noted that this improvement is obtained at the expense of spatial resolution. The improvement may be less pronounced when topographic features are used because of their lower scattering efficiencies relative to a retroreflector.

The range over which an atmospheric backscatter DIAL system operates depends on the detector configuration. For DIAL systems operating in the infrared spectral region, the range is limited to 1 to 2 km when direct detection is used. The region can be extended to 5 to 8 km when heterodyne detection is used.

A basic DIAL system is presented in Figure 4. The wavelength of Laser 2 is associated with the absorption feature of the target species and the wavelength of Laser 1 is not. When the target species is not present in the atmosphere, the signals reaching the receiver from the two lasers will be equal. When the target species is present, Signal 2 will be less than Signal 1. The ratio of the two signal intensities is a function of the amount of the target species that is present.

By allowing the laser sources and the receiver optics to scan through a volume, a three-dimensional map of concentration can be generated. For long pathlengths, high-powered lasers or sensitive optical detection techniques must be employed because of the weak reflected signal strength. For pathlengths less than 250 m, low-powered, continuous wave lasers and pyroelectric detectors may be used (O. A. Simpson, Environmental Laser Systems, personal communication, 1988). Applications to emissions monitoring include the measurement of sulfur dioxide and nitrogen dioxide from point sources, nitrogen



R_1
Laser 1
M
BS
Laser 2
BS
R_2
M
Gas
Cube corner
retroreflector
or background
Receiver
No gas
present
Signal 1
Signal 2
Low
quantity
of gas
present
Signal 1
Signal 2
High
quantity
of gas
present
Signal 1
Signal 2

BS = Beamsplitter
M = Mirror
R_1, R_2 = Laser power reference signal

Figure 4. Basic DIAL lidar system configuration.[45]

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dioxide from line sources or area sources, and other species emissions for which appropriate laser frequencies can be produced. These applications are discussed in Section 3.

Fluorescence lidar--Fluorescence lidar is an active remote sensing technique for monitoring minor species in the atmosphere.[46] It is based on detection of the fluorescence of atoms and molecules that have been excited with a resonance-tuned probe laser. The basic idea behind fluorescence lidar is quite simple. A pulsed laser, which is tuned to an absorption line of the species to be studied, is used to generate a probe beam that is projected into the atmosphere. The probe beam excites a calculable fraction of the target species into excited states. These target species subsequently fluoresce with a known quantum yield. The resulting fluorescence is detected by a receiver located at the same position as the probe laser. Because the laser is pulsed, the fluorescence that is detected at some specific time after the pulse can be associated with some specific distance from the lidar. The intensity of the fluorescence that is detected is a function of the distance and the density of the species at that distance. The technique can obtain density information with a range up to 100 km or more with spatial resolutions to tens of meters.

The major constraint on fluorescence lidar is quenching. The pressures involved in the troposphere and stratosphere are sufficient to lead to multiple molecular collisions during the time between a molecule's excitation and its subsequent fluorescence. The collisions result both in quenching and in energy redistribution. Quenching removes energy from the excited state without fluorescence and thereby reduces the total quantum yield. The redistribution of energy from the desired excited state into other excited states results in fluorescence at red-shifted wavelengths. The redistribution can reduce the effective quantum yield into the detector bandwidth. Alternately, it can be used to discriminate against Rayleigh or aerosol scattering, which is not spectrally shifted. It is the quenching that makes fluorescence lidar a difficult and inefficient technique for monitoring species in the atmosphere.

Another constraint on fluorescence lidar is solar background radiation, which generally reduces the technique's applicability to nighttime measurements. When a fluorescence lidar is used during the daytime, the major source of noise is usually scattered sunlight. If an atomic or simple diatomic molecule is being probed, the choice of a detector bandwidth for daytime measurements often comes down to using as monochromatic a bandwidth as possible, while still overlapping the few fluorescent lines. When a more complicated and extensive fluorescence spectrum (such as that for nitrogen dioxide) is involved, it is necessary to consider in detail the spectra of the fluorescence and the scattered sunlight to determine the optimum bandwidth for the detector.

Species that can be studied with fluorescence lidar are those that can be excited to fluoresce in the 200 to 1,000-nm spectral region. Molecular absorption cross-sections are higher in this region than in the infrared region. Additionally, radiative lifetimes for excited states are shorter in this region, which reduces the changes for quenching and energy distribution via molecular collisions.

The most successful application of fluorescence lidar to date has been the detection of sodium and potassium atoms in the upper atmosphere from an altitude of 80 to 100 km.[47,48] These measurements were made during the night. In 1980, experiments were underway by NASA's Goddard Space Flight Center (NASA/Goddard) and the University of Maryland to use a balloon-borne fluorescence lidar to measure the stratospheric hydroxyl radical density.[46] Calculations showed that nitrogen dioxide measurements were feasible, especially at night and at limited ranges. The greater difficulty in measuring the hydroxyl and nitrogen dioxide densities can be illustrated by comparing the following absorption cross sections: sodium at 1.8×10^{-11} cm^2; hydroxyl radical at 6.2×10^{-15} cm^2; and nitrogen dioxide at 1.6×10^{-21} cm^2.

It is doubtful that fluorescence lidar will have any application to measurements of atmospheric emissions from stationary sources.

Raman backscatter lidar--When light passes through a liquid or gas, a small fraction of the light is scattered at the same wavelength as the incident light.[49] This Rayleigh scattering may be viewed as the elastic scattering of a photon by a molecule. Sometimes weak radiation at wavelengths longer than those of the incident light is also present in the scattered light. This Raman scattering may be viewed as the inelastic scattering of a photon by a molecule. The wavelength differences between the weak lines and the exciting line are characteristic of the scattering molecule and are independent of the wavelength of the exciting line.

The usefulness of Raman scattering is substantially restricted to the optical and near-optical wavelengths between 100 and 10,000 nm.[50] In addition to the wavelength shift associated with Raman scattering, a second very important characteristic is the size of the Raman cross-section. It is significantly smaller than that for the absorption process. In the most extreme cases, the absorption cross-section can exceed the Raman cross-section by as much as 20 orders of magnitude. More typically, for a variety of reasons, practical differences of five to six orders of magnitude are common in atmospheric probing. Prescinding from resonance scattering, the Raman cross-sections range from 10^{-26} to 10^{-32} cm^2, while the absorption cross-sections range from 10^{-18} to 10^{-24} cm^2. Therefore, high-intensity lasers operating in the visible or ultraviolet wavelengths are required for Raman lidar.

Raman lidar is generally limited to clear-sky, nighttime applications because of noise that arises from background solar radiation. However, daylight applications are possible if the Raman lidar is operated in the solar-blind spectral region (230 to 310 nm).[50] In this region, the ambient solar flux is essentially reduced to zero because of absorption by stratospheric ozone.

Raman lidar has been most frequently used for measurements of atmospheric water vapor profiles.[50,51] Detailed, reliable profiles were obtained up to a height of 2 to 3 km. Other applications have included measurements of methane plumes from simulated liquid natural gas spills at a range of 300 m,[51] subsurface ocean temperature profiles[51] and sulfur dioxide, carbon dioxide, and kerosene measurements at a range of 200 meters.[52,53] These latter measurements had a 10-m range resolution. They were taken in full daylight under both clear atmosphere and fog conditions. Good signal-to-noise ratios were obtained with sensitivities of 300 ppm-m for sulfur dioxide and 17 ppm-m for kerosene.

Doppler lidar--Many EPA emission standards are based on the mass emission rate of the pollutant of interest. This rate is a function of the pollutant concentration and the plume velocity. The other methods that are described in this report measure pollutant concentration. Doppler lidar provides a means to obtain information about gas velocity. This technique has been developed extensively for tropospheric wind field measurements from airborne and orbiting platforms and for in-stack velocity measurements. It has also been used for remote measurements of plume velocity.

A general overview of atmospheric laser Doppler velocimetry has been given by Bilbro.[54] The underlying principle of operation for laser Doppler velocimeters rests in the fact that electromagnetic radiation passing through a medium is scattered by the inhomogeneities that are within that medium. The primary scatterer is naturally entrained particulate matter such as pollens, dust, and various pollutants whose diameters are on the order of from 1 to 10 μm. The majority of these naturally occurring aerosols tend to follow the motion of the atmosphere in which they are entrained. Hence, the velocity of these particles provided a reliable measure of the velocity of the surrounding wind field.

The illumination of particles by a laser source results in multidirectional scattering. Some of the incident radiation is backscattered along the axis of illumination. One property of the backscattered radiation concerns the Doppler effect, in that the backscattered radiation has been shifted in frequency from the incident radiation by an amount proportional to the particle velocity component parallel to the axis of illumination (radial velocity). The extraction of the Doppler shift from the backscattered radiation is accomplished by coherent mixing (heterodyning) of the backscattered radiation with radiation from a reference local oscillator. When the two beams are mixed, an interference pattern is created that is imaged on a photodetector. The interference pattern fluctuates according to the frequency difference between the two beams. This fluctuation produces a detector output in the form of a frequency-modulated signal whose modulation frequency is the Doppler-shifted frequency.

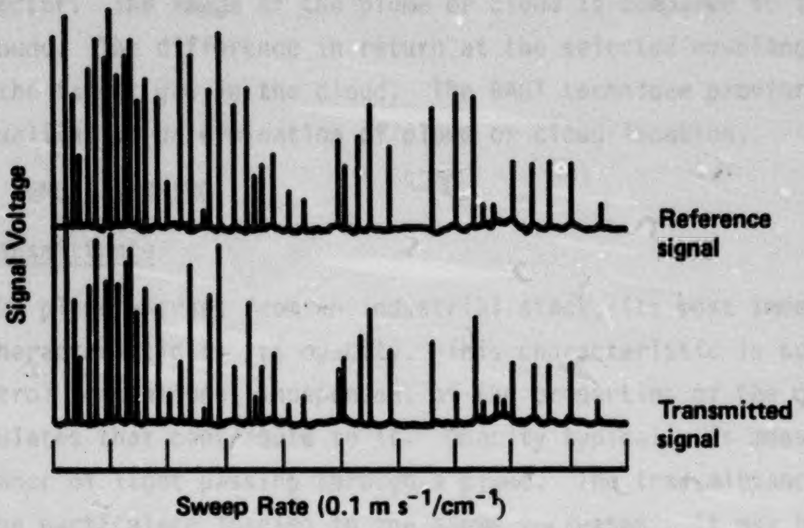
U.S. EPA sponsored the development by Raytheon Corporation (Sudbury, Massachusetts) of a mobile laser-Doppler velocimeter that would be dedicated to air pollution measurements.[55,56] This system consists of the following major subsystems: an optical subsystem, a scan and pointing subsystem, a signal processing subsystem, and a data recording subsystem. The optical portion of the system includes a 5-W air-cooled carbon dioxide laser, a Mach-Zehnder interferometer, a telescope, a two-axis output mirror for pointing the laser's beam, and a liquid-nitrogen-cooled mercury-cadmium-telluride detector. A portion of the laser's light is directed through the interferometer to the detector to serve as a reference signal. The remainder of the light is directed through the interferometer, the telescope, and the output mirror. Backscattered and Doppler-shifted light from the plume returns through these same optical components to the detector. By comparing information contained in the reference and backscatter signals, the signal processing subsystem determines the gas velocity in the plume.

U.S. EPA has conducted preliminary measurements of stack exit velocity at a coal-burning power plant using this system. A van housing the system was located 300 or 400 m from the stack exit. As the Doppler lidar measured the stack exit velocity, in-stack pitot-tube measurements of the stack effluent velocity were made using portholes at the base of the stack. After correcting for the difference in the areas of the base and the exits, the two sets of measurements agreed to within approximately 14 percent. An error in the area correction factor was thought to be the reason for the difference between the Doppler lidar and in-stack measurements.

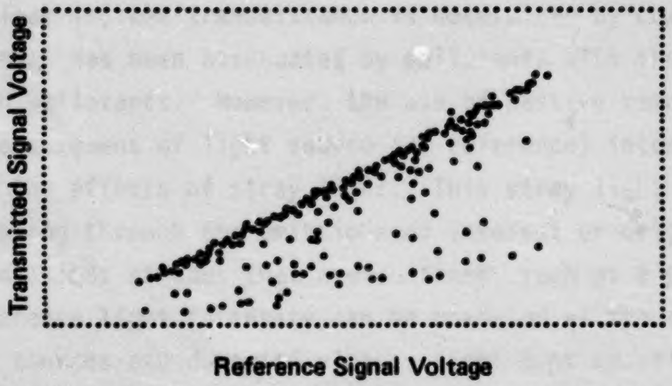
Wedge absorption lidar--Another technology applicable to pollutant sources is wedge absorption lidar.[57,58] This technique employs a solid-state erbium:ytterbium-aluminum-garnet (Er:YAG) laser operating in long-pulse mode. The operation of the solid-state laser produces emission lines that cover a range of frequencies during a single pulse. Some of the emission lines for selected solid-state lasers correspond to absorption lines of selected trace atmospheric species. The system operates similarly to DIAL, except that the on and off absorption information is obtained during a single pulse of a single laser and no tuning is required. A plot of transmitted signal power versus reference signal power produces a wedge. An example is shown in Figure 5. The upper slope represents lines that are not absorbed, and the lower slope is absorption lines of the species measured. The ratio of the lower and upper slopes of the wedge is proportional to the concentration of the absorbing gas. The technique is limited by the rare occurrence of absorption line coincidences. Methane can be measured using an erbium laser, and carbon dioxide can be measured with a holmium laser.

Gas Imaging--

Backscatter/absorption gas imaging (BAGI) is a technique by which invisible source vapor plumes or clouds are displayed as a visible signal on a standard television monitor. A carbon dioxide laser serves as the light



a. Dual beam oscilloscope trace of the reference (top) and transmitted (bottom) long-pulse spike train of an Er:YAG laser (y-coordinate is 2 V/cm and x-coordinate is 0.1 ms/cm.)



b. Cartesian coordinate display of absorption wedge representation of data from a few Er:YAG "long" pulses (relaxation pulse trains) propagated 480 m through a 760 torr total pressure atmosphere containing 0.018 torr of methane.

Figure 5. Example of wedge absorption data representation.[57]

source. The laser is directed to the location of the emissions. The target gas species absorbs characteristic wavelengths, and the radiation is reflected back to a detector. The image of the plume or cloud is compared to the image of the background. The difference in return at the selected wavelengths is dependent on the target gas in the cloud. The BAGI technique provides a real-time visual qualitative determination of plume or cloud location.

PRINCIPLES OF REMOTE SENSING

Opacity or Transmittance

As a smoke plume emerges from an industrial stack, its most immediately perceptible characteristic is its opacity. This characteristic is subject to emissions control regulations, independent of the properties of the gases and/or particulates that contribute to it. Opacity typically is measured by the transmittance of light passing through a plume. The transmittance will decrease as the particulate loading in the plume increases. It may be measured by in-stack instruments or remote sensors. In-stack instruments are discussed in detail in Reference 59. The remainder of this discussion deals with remote sensors.

The principle upon which remote sensing devices are based is the same as that employed for the standard laboratory spectrophotometer or in-stack transmissometers. That is, the transmittance is determined by comparing the intensity of light that has been attenuated by pollutants with the intensity in the absence of such pollutants. However, the use of passive remote devices complicates the measurement of light source (or reference) intensity and the elimination of the effects of stray light. This stray light enters the detector without passing through the emissions of interest or originating from the source. For emissions streams that are confined, such as a plume near a stack, the reference light intensity can be measured at the side of the plume. For large area sources and downwind plumes, light must travel great distances to be measured by remote sensing devices. Error may be introduced by changes in reference light intensity or travel time. Temporal changes in the source intensity can be accounted for by using two detectors that are calibrated to one another. One detector continuously views the radiation source directly outside the emission stream of interest while the signal detector continuously views the source through the emission stream.

The components of radiation from a source and components of stray light entering a detector are illustrated in Figure 6.[60] Rays a, b, and c originate from the source, but only c enters the detector. The intensity of ray c may be decreased because of absorption by the gas in the plume depicted at the left. Ray a represents radiation that is scattered by the particulate matter in the plume. Ray b represents radiation that is scattered or absorbed between the emission stream and the detector, thus reducing the effective signal. Rays d, e, and f represent radiation that enters the detector but does not originate from the primary light source, thus causing an offset in the signal. When transmittance of the emission stream is measured, all of these rays are nonideal except a and c. When analysis is conducted for gaseous emissions, all of these rays are nonideal except c. When analysis is conducted for particulate emissions, the change in intensity of ray c (due to the emitted gases) is a nonideal factor.

Determination of the source intensity is depicted at the right of Figure 6. If the procedure is such that ray b is the same between measurement of the transmitted radiation and the source radiation, and ray c is sufficiently intense, then ray b produces no error. Ray f produces a positive error (high transmittance) in the measurements. Its effects can be minimized by ensuring that it is small relative to the source intensity, ray c. Note that conditions such as haze and long distances between the emissions and the detector tend to increase f while decreasing c. Radiation, represented by rays d and e, which also produces positive error, is due to scattering of ambient light into the view of the detector by particulate matter in the emission streams. For techniques in which the primary source is not the sun, this type of interference is minimized by positioning the detector so that the sun is behind it. Still, some techniques are limited to characterizing emissions with low levels of particulate matter because of this radiation. Designers of passive remote sensors have attempted to incorporate features that distinguish this type of stray light from that originating from the primary sources. These schemes will be described in conjunction with each device.

One approach is the sun photometer, which views the sun through and to one side of the emissions. This source then dominates all stray light (on clear days). These devices are limited because of the requirement to change location frequently; however, if the emissions to be detected are spread over

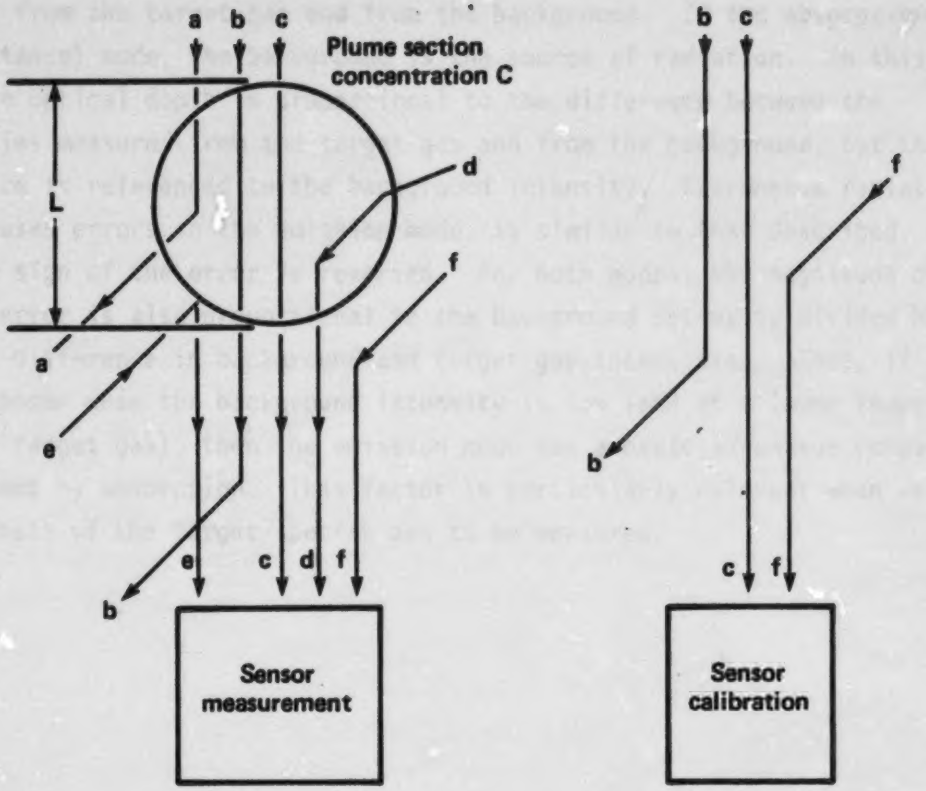

a
b
c
Plume section concentration C
L
d
f
a
e
b
e
c
d
f
Sensor measurement
b
c
f
b
c
f
Sensor calibration

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Figure 6. Components of radiation involved in passive remote measurement of emission.[60]

a broad area, simple tracking of the sun may be accomplished by rotating the detector to keep the pollutants in the detector view.

Absorption Mode Versus Emission Mode

For passive techniques in the emission mode, the target gas is the source of radiation, and the optical depth (i.e., the product of concentration times pathlength) is proportional to the absolute difference between intensities measured from the target gas and from the background. In the absorption (or transmittance) mode, the background is the source of radiation. In this mode, also, the optical depth is proportional to the difference between the intensities measured from the target gas and from the background, but this difference is referenced to the background intensity. Extraneous radiation, which causes errors in the emission mode, is similar to that described, except that the sign of the error is reversed. For both modes, the magnitude of percent error is also proportional to the background intensity divided by the absolute difference in background and target gas intensities. Thus, if a time can be chosen when the background intensity is low (and at a lower temperature than the target gas), then the emission mode has a basic advantage compared to measurement by absorption. This factor is particularly relevant when very small levels of the target species are to be measured.

SECTION 3

SUMMARY OF REMOTE SENSING TECHNOLOGIES

COMMERCIALLY AVAILABLE INSTRUMENTS

Passive Techniques

Radiometers--

Visiplume--A recently developed radiometer, the Visiplume Model 121 sulfur dioxide/velocity/opacity monitor, employs an ultraviolet-sensitive video system (vidicon camera) to monitor and display sulfur dioxide emissions in a plume.[61] The source of radiation is skylight. The system uses spectral bandpass filters to isolate the sulfur dioxide absorption band around 300 nm and a slightly longer wavelength out of the sulfur dioxide absorption region. Transmittances, measured at the two wavelengths, are compared to eliminate the effects of particulate scattering of source radiation. As illustrated in Figure 7, the video signal corresponds to the locations of the two cursors, one for the reference intensity and one for the transmitted intensity. The instrument is also designed to measure velocity of the stream of emissions so that the total volume flow rate is determined.

The instrument was evaluated and found to meet the manufacturer-claimed limits of ±150 ppm in 31 of 36 different test conditions in the sulfur dioxide concentration range of 0 to 2,215 ppm. The relationship between the Visiplume and a reference method is shown in Figure 8. This comparison incorporates data obtained from smoke generator tests, bag tests, and field tests. The correlation between the two measurement techniques was good. The bag test data were taken over a 0 to 2,215 ppm range. Linear regression of 10 tests produced a slope of 1.00 with a correlation coefficient of 0.989 and a 2-σ standard error range of ± 218 ppm. The smoke generator data were taken over a 0 to 1,614 ppm range. Linear regression of 13 tests produced a slope of 1.22 with a correlation coefficient of 0.997 and a 2-σ standard error range of ± 116 ppm. Field test data acquired at two sites were taken over a 0 to 471 ppm range. Linear regression of 13 tests produced a slope of 0.98 with a correla-

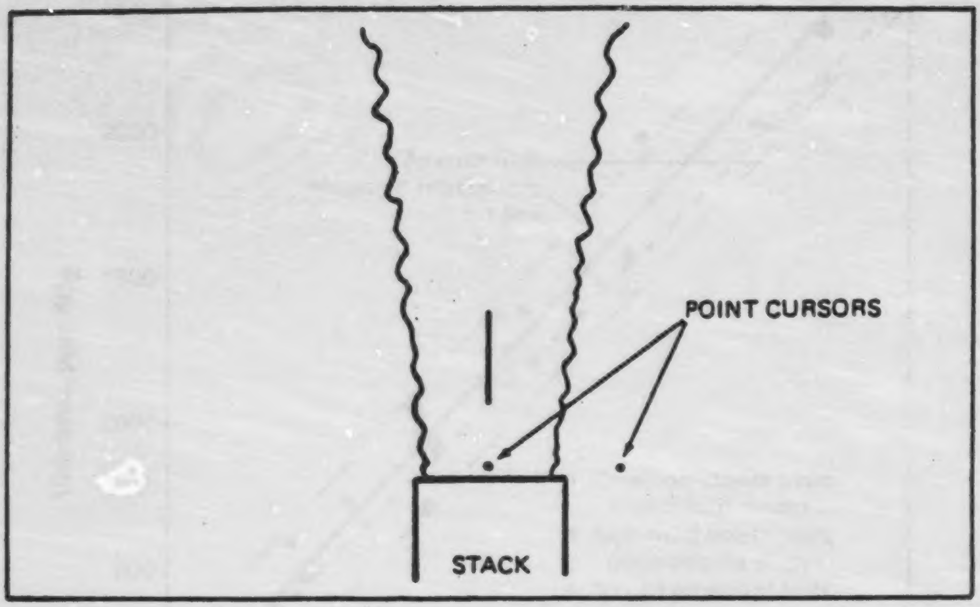

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Figure 7. Correct cursor pattern position for plume measurement with a radiometer.[61]

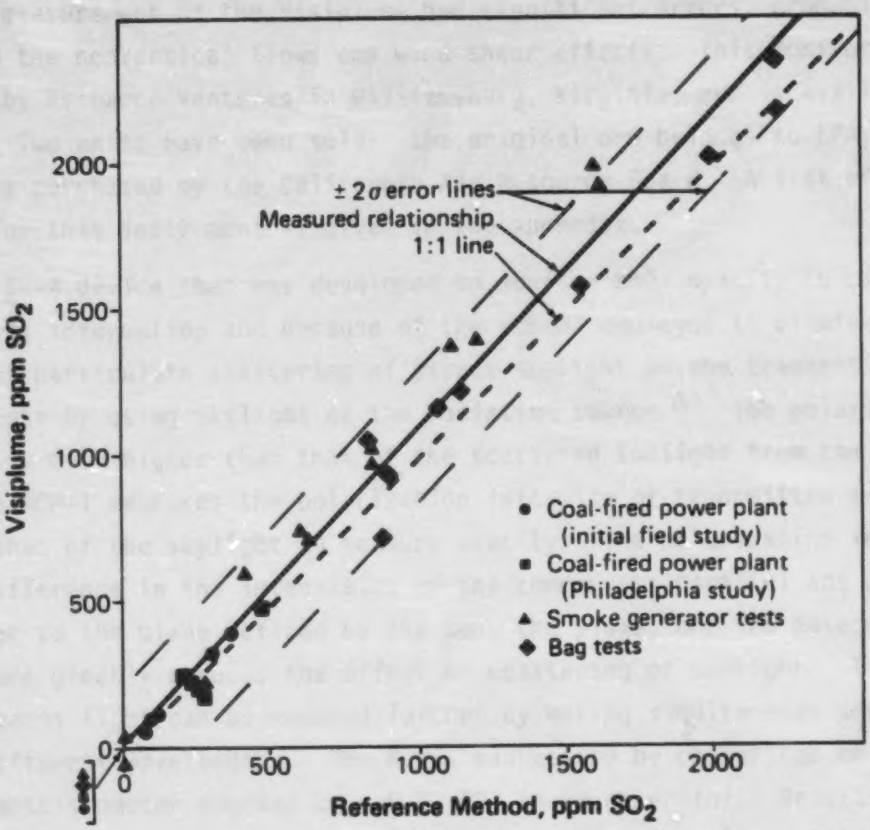

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Figure 8. Sulfur dioxide comparative measurements by visiplume and reference methods.[61]

tion coefficient of 0.974 and a 2-σ standard error range of $\pm$66 ppm. In Figure 9, the regression lines from the three sample types are shown separately. The Visiplume had a positive bias of about 20 percent in the smoke generator tests, but the agreement to the reference method was within 3 percent for the bag tests and field tests. The Visiplume response was immune to effects of extraneous light (sunlight scattered by the plume) for particulate loadings up to levels described as approximately 22 percent opacity. The velocity measurement of the Visiplume had significant errors, probably associated with the nonvertical flows and wind shear effects. This monitor is manufactured by Research Ventures in Williamsburg, Virginia, and is available for $37,000. Two units have been sold: the original one belongs to EPA, and the second was purchased by the California Air Resource Board. A list of specifications for this instrument is given in the appendix.

ROM-I--A device that was developed to monitor only opacity is of interest for general information and because of the scheme employed to eliminate the effects of particulate scattering of direct sunlight on the transmittance measurements by using skylight as the radiation source.[62] The polarization of skylight is much higher than that of the scattered sunlight from the plume. Thus, the ROM-I measures the polarization intensity of transmitted light relative to that of the skylight to measure opacity. The polarization intensity is the difference in the intensities of the components parallel and perpendicular to the plane defined by the sun, the plume, and the detector. This measurement greatly reduces the effect of scattering of sunlight. The effects of extraneous light can be reduced further by making simultaneous measurements at two different wavelengths. The ROM-I was tested by comparison to an in-stack transmissometer mounted on a U.S. EPA smoke generator. Results are given in Figure 10. For varying solar angles (time of day) and different days, the correlation coefficient was always greater than 0.96. The ROM-I is available commercially from GCA Corporation, Bedford, Maine, at a cost of $7,000. One has been sold, and several have been constructed.

Plumetracker--Moniteq Ltd. of Concord, Ontario, Canada, has designed a remote sensor for the simultaneous measurement of sulfur oxide and nitrogen dioxide. The instrument's principle of measurement is differential ultraviolet absorption. Its main components are a light-gathering telescope, a

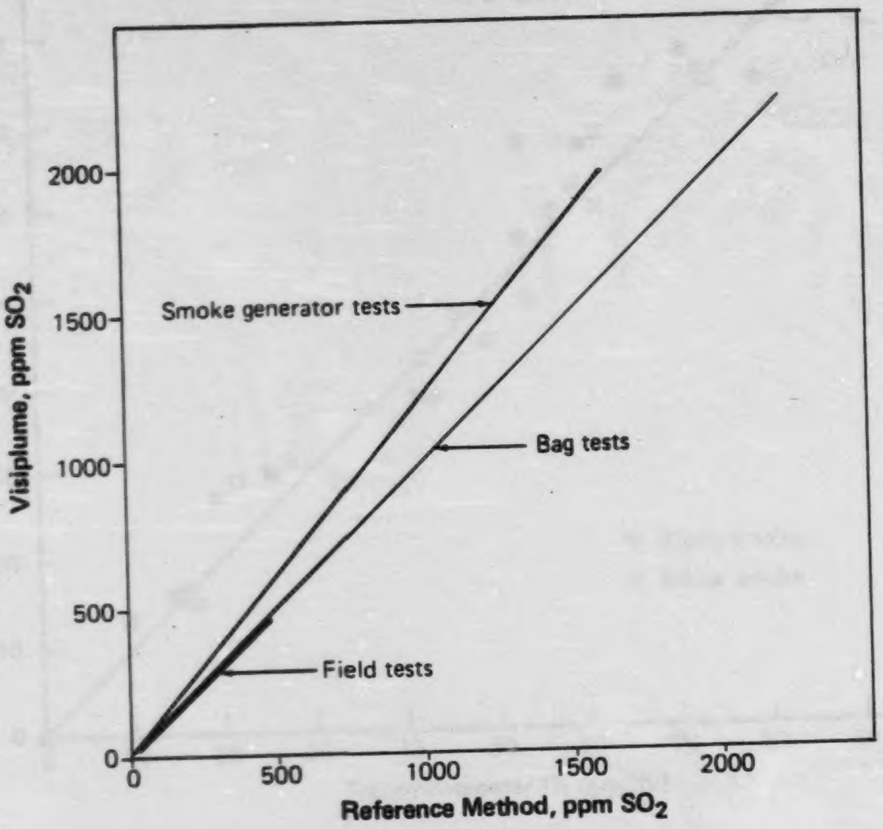

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Figure 9. Linear regression analysis for each major test type.[61]

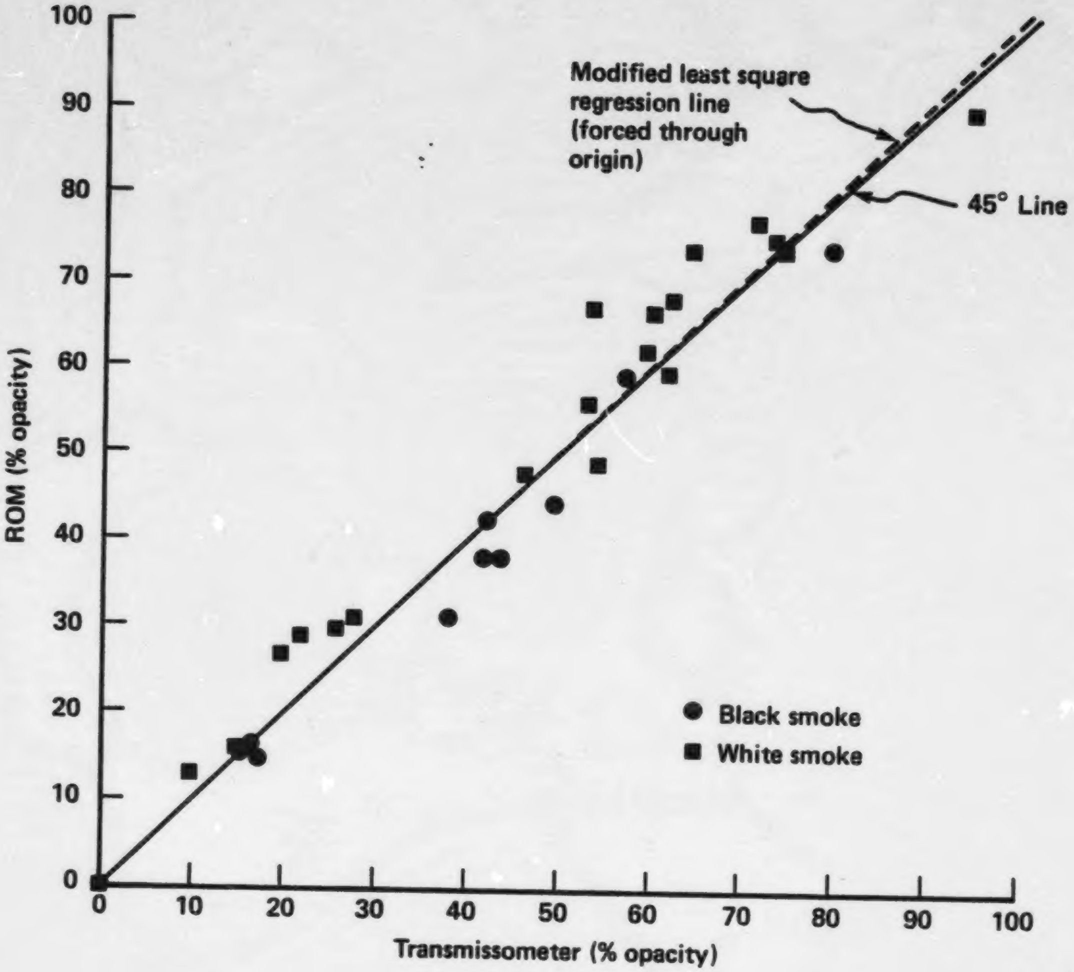

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Figure 10. ROM-I versus transmissometer data for a smoke generator plume.[62]

grating spectrometer with multiple exit slits, two detectors, and signal processing electronics. Ultraviolet sunlight that has passed through a plume enters the spectrometer. Exit slits select the wavelengths of the two target gases to be measured. Two exit slits per gas are arranged to monitor an absorption spectrum maximum and its adjacent minimum. A rotating aperture mask alternately passes the light from each slit into a photomultiplier tube. The instrument can be used either as an upward-looking sensor mounted in a vehicle or as a downward-looking sensor mounted in a helicopter. It weighs 16 kg. Its power requirements are 150 W at any one of several line voltages. It has a detection limit of 1 ppm-m for both sulfur dioxide and nitrogen dioxide. It has been used for mass emission measurements in Canada, Australia, Czechoslovakia, and South Africa.

The instrument's distributor in the United States is Recon Exploration of Dallas, Texas. Its current price is $50,000 in this country. A list of specifications for this instrument is given in the appendix.

Mask Correlation Spectrometers--

COSPEC--This is the best known instrument incorporating mask correlation spectroscopy and was developed by Barringer Research, Ltd., Rexdale, Ontario. The COSPEC instrument has been used to perform an extensive number of field measurements for sulfur dioxide and nitrogen dioxide since 1971.[63] In the active mode, the instrument uses chopped xenon or quartz iodine lamps for long-path measurements. Results of these tests, which were compared to results using wet chemistry techniques, indicate COSPEC sensitivities of 2 ppb over a 300-m path. The actual peak-to-peak noise resolution of the instrument, however, was closer to 15 ppb integrated over the 300-m path. In the passive mode, baseline drift, caused by the changing source spectra, is an added variable. This limits the accuracy to 40 ppm-m over several hours during the middle of the day[64] and 100 ppm-m over an entire day.[65] The instrument gives a linear response for 0 to 2,000 ppm-m but can be modified to extend the range to 10,000 ppm-m.[66] The accuracy for determining the total flux of sulfur dioxide plumes over long distances has been estimated to be less than 20 percent, and essentially all of that error is due to the inability to measure air volume flow.[66] Results of one study, in which three COSPECs were used to estimate sulfur dioxide emission rates from a coal-fired power station, were within 7 percent of those computed by the utility.[67] In other studies, two COSPECs were used to locate a plume during a period of rain,[68] and a COSPEC detected the volcanic eruption cloud from Mt. St. Helens over Ontario 2 days after the eruption.[69]

An extensive field program was undertaken by the Commission of European Communities (CEC) in 1975 to evaluate remote monitors of sulfur dioxide.[70] The site chosen was the Gave de Pau Valley in France where a plant that produces and desulfurizes natural gas is located. Measurements of the plume from this plant over long distances were performed. In-stack monitors provided actual total emissions of sulfur dioxide. In one set of tests, three COSPECs were compared during traverses under the plume (monitors facing upward). Measurements by two of these instruments agreed within 10 percent, whereas the sulfur dioxide value from the third was 20 percent lower. The variance of total emissions for a single traverse was 30 percent, and the average value from the COSPECs was 80 percent higher than the values from the in-stack monitors. Most of this error was attributed to uncertainties in measurement of plume velocities rather than error in measurement of optical depth. Comparison of the COSPECs to the selective modulation radiometer (described later in this section) showed good agreement, but no quantitative information was reported. The COSPECs in the active mode agreed within 10 percent to the TELETEC (described below).

The COSPEC has been used for remote sensing of sulfur dioxide in plumes close to the stack. In one study, the COSPEC monitored two stacks at a large thermal generating station and an oil refinery stack.[60] Measurement had to be made at three distances away from the stack to account for extraneous light. When extrapolated to the stack, the data for the power station indicated concentrations of 1,200 and 1,300 ppm, which compared favorably with a wet chemistry value of 1,430 ppm. Values from the oil refinery stack also agreed with wet chemistry results to 20 percent.

The basic COSPEC V system costs $35,000 (C. Cumming, Barringer Research, Ltd., Rexdale, Ontario, personal communication, 1987) and complete computerized system to be mounted in a station wagon for plume chasing costs $65,000. The quoted sensitivity was 2 to 5 ppm-m with a clear sky and 6 h of operation during the brightest part of the day for sulfur dioxide and 11 h for nitrogen dioxide. The quoted accuracy was 5 percent of the reading. One instrument can monitor sulfur dioxide and nitrogen dioxide, but 1 h is required to convert from one gas to the other. Approximately 100 COSPECs have been sold. For monitoring close to a stack, the method of measuring at several distances and then extrapolating can be avoided by installing a larger telescope on the COSPEC to obtain a smaller field of view without losing sensitivity. However, this telescope was estimated to cost an additional $50,000. Lists of specifications for two Barringer mask correlation instruments are given in the appendix.

TELETEC--The TELETEC is an instrument fashioned after the COSPEC in its active mode and commercialized by Tecneco (Fano, Italy) to extend useful range to several kilometers.[24] One variation from the COSPEC is the incorporation of a gas calibration cell that can cover a large range of optical depths without changing the sample gas content. This is accomplished with an extensible tube that elongates without changing its volume. The other modification was in the method for obtaining the optical path to include the first derivative of the detector signal with respect to optical depth, which is measured onsite with the extensible tube. These modifications made the technique more immune to environmental conditions affecting the spectral makeup of the radiation entering the instrument, which changes in propagating long distances through the atmosphere. Another improvement[25] in this technology, also adopted for the COSPEC, was the conversion from a reciprocating mask to a circular disk with grooves rotating at high speed. This advancement, with faster electronics, reduced the time required for a single measurement from 5 to 20 min to 10 to 20 s. Thus, the effects of atmospheric scintillation were eliminated, and rapid changes in plumes could be monitored. It appears that the instrumental noise was reduced from ±70 percent to ±10 percent.

Gas Correlation Spectrometers--

GASPILS--Moniteq Ltd. of Concord, Ontario, Canada, has designed an airborne remote sensor for the detection of methane leaks from gas pipelines.[71] This instrument differs from other gas correlation spectrometers because it measures two components rather than a single component. Because the second component (e.g., ethane or nitrous oxide) is measured, the methane measurements can be corrected for background methane and interferents in the atmosphere and for temperature and emissivity variations in the ground. The detection limit for methane is less than 20 ppm-m at a wavelength of 7.8 μm and a temperature differential of 4 °C. Many other compounds can be measured with this instrument. The instrument is aircraft-compatible and weighs 18 kg. Its power requirements are 165 W at startup and 100 W while in operation using a supply voltage of 28 V dc.

The instrument's distributor in the United States is Recon Exploration of Dallas, Texas. Its current price is $150,000 in this country. A list of specifications for this instrument is given in the appendix.

GASPEC--This gas correlation spectrometer can be purchased from Barringer Research, Ltd., which developed the instrument to measure tropospheric carbon monoxide from the space shuttle.[27,72] Several of these instruments have been built and tested and have been configured to measure methane, ethane, hydrogen chloride, and carbon monoxide. Discussions (C. Cumming, Barringer Research, Ltd., personal communication, 1987) indicated that a GASPEC for other target gases with infrared absorption spectra could be developed for between $75,000 and $100,000. A list of specifications for this instrument is given in the appendix.

Interferometers--

ROSE--A mobile remote interferometric sensor was developed by U.S. EPA in the late 1970s.[12] Although the technique looked promising, the system has been disassembled. This system consisted of a Nicolet 7199 Fourier transform infrared (FTIR) system configured to fit into a van. This remote optical sensing of emissions (ROSE) system could be operated in three modes, one of which was passive. The passive mode was based on single-ended emission measurements. It could also be used as an active sensor in either a long-path absorbance (up to 2 km) mode by using a remotely located quartz-halogen lamp or in a cross-stack absorbance mode.[73-75] The optical components of the system are illustrated in Figure 11. The instrument's spectral resolution is 0.125 cm^{-1}. Its spectral range is from 700 to 1,800 cm^{-1} using a mercury-cadmium-telluride detector and from 1,800 to 6,000 cm^{-1} using an indium-antimonide detector. The nominal sensitivity of the ROSE system for many pollutant gases was about 1 ppm-m for active absorption measurements and close to 10 ppm-m for emission measurements.[73] The accuracy of the active absorption measurements was less than 10 percent. The error in emission measure-

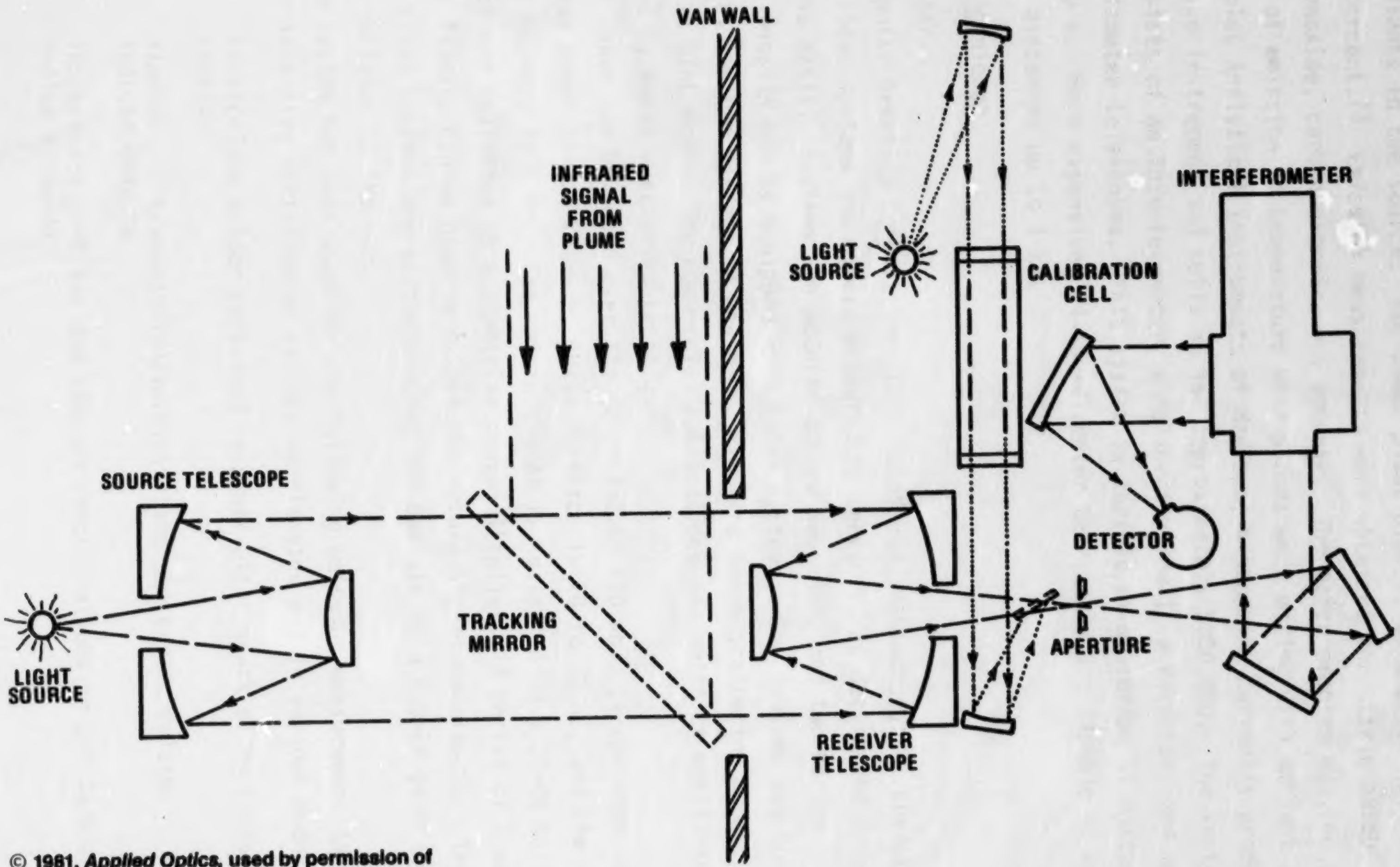
VAN WALL
INFRARED SIGNAL FROM PLUME
LIGHT SOURCE
CALIBRATION CELL
INTERFEROMETER
DETECTOR
SOURCE TELESCOPE
TRACKING MIRROR
LIGHT SOURCE
APERTURE
RECEIVER TELESCOPE

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Figure 11. EPA ROSE Infrared spectrometer system.[12]

ments can be largely attributed to uncertainty in temperature and concentration gradients in the source. For power plant plumes, this error was less than 20 percent.[73] Emission measurements were obtained for nitric oxide, carbon monoxide, carbon dioxide, and ammonia. The major problem was the sensitivity of emission to temperature when plumes were mixing with ambient air.

Nicolet Analytical Instruments of Madison, Wisconsin, currently produces a ROSE-like instrument and sells it for approximately $100,000. The instrument consists of an interferometer, a calibration cell, a detector, and two 14-in. diameter telescopes. This system is capable of measuring at distances up to 500 m. More expensive, 24-in.-diameter telescopes are capable of measuring at distances up to 1 km.

Active Techniques

Raman Lidar--

Computer Genetics Corporation (CGC; Wakefield, Massachusetts) operates several lidar systems and offers measurement services, as determined by need, on a lease basis. Systems are mounted aboard two vans, and two marine research vessels can be equipped with lidar systems. The systems use tunable dye lasers and can operate in Rayleigh, Mie, and Raman scattering modes, as well as in DIAL mode. The commercial applications for emission monitoring are performed by Raman scattering techniques.

The laser can be tuned over the range 350 to 750 nm. It operates at a pulse peak power of 4.5×10^6 W; pulse duration is 10 to 20 ns, and the repetition frequency is 8 Hz. The return signal is collected in a 30-cm coaxial telescope and delivered to a sensitive photomultiplier. A series of 1-nm bandpass filters can be used to select the return signal wavelength. The van-mounted lidar systems are self-contained and operate on a 6.5-kW generator that is included in the van.

The system has been used for the following emission measurement applications by selective operation at various wavelengths and in various modes:

- Particulate matter contained in power plant stack plume cross sections
- Atmospheric transmission/turbidity and smoke/particulate-induced opacity
- Temperature profiles and species concentrations in jet turbine engine exhausts

- Range-resolved methane concentration levels in dispersing liquid natural gas spill clouds

- Tracking of atmospheric dispersed dye tracers such as rhodamine.

Systems can be configured with varying sophistication to achieve sensitivities down to parts per billion levels at distances of over 1 km. A list of specifications is given in the appendix to this report.

The cost of building a van-mounted system would be between $400,000 and $700,000, depending on the specifications desired. CGC leases their van-mounted systems for measurement applications at a basic rate of $30,000 per month, plus a transport charge of approximately $1.00/km. The basic cost for an operator and data reduction services is $35,595 per month. A list of specifications for this instrument is given in the appendix.

Differential Absorption Lidar--

Environmental Laser Systems, Incorporated (ELS; Atlanta, Georgia), offers a versatile carbon dioxide laser system that operates on the DIAL principle. The system is known as LASERSAFE, which is an acronym for laser remote sensing of atmospheric fugitive emissions. LASERSAFE can be configured for continuous-wave scanning mode or pulsed, line-of-sight mode. In continuous scanning mode, the system can scan an area 50 m x 50 m at a range of 200 m. Operating distance is 100 m for topographic targets and greater than 500 m using retroreflectors. In pulsed operation, an area 280 m x 280 m can be scanned at a 1-km distance. The system can be operated over a distance of 5 km in line-of-sight, pulsed mode by using a retroreflector. Alternative configurations can be employed, depending on the application and species to be measured.

The system is operated on 110 V ac. It weighs approximately 140 kg, and therefore is mounted in a van for mobile applications. The sensitivity varies by species measured but is typically between 1 and 400 ppm-m. Response time is less than 1 s, and accuracy is specified as typically better than 5 percent. A list of species that have been measured with LASERSAFE and a copy of the manufacturer's specifications sheet are included in the appendix.

Typical applications of LASERSAFE are

- Chemical plant and petroleum refinery scanning

- Off-shore drilling platform surveillance

- Oil and gas pipeline leak detection
- Environmental surveys
- Perimeter monitoring
- Hazardous waste disposal site surveillance
- Storage tank area scanning.

System costs are $125,000 to $180,000, depending on the features selected and complexity of the system. ELS has outfitted a van with a LASERSAFE system and provides measurement services for short periods or in other applications where the cost of purchasing the instrument is not justified.

Long-Path Infrared Absorption--

Wright and Wright, Incorporated (Oak Bluffs, Massachusetts), markets a series of long-path infrared hydrocarbon vapor monitors.[76] Additionally, their instruments have been modified for measurements of hydrogen fluoride and, potentially, for hydrogen sulfide. Each of the separate systems is based on a long-path principle but uses the differential transmission of two distinct wavelengths to detect the gas concentration. The light source is a quartz halogen lamp. The dual wavelength detection is accomplished by rotating a filter wheel in front of the infrared photodetector. The rotating filter wheel effectively chops the polychromatic light source and passes a reference wavelength and the hydrocarbon-specific wavelength to the photodetector. Signal-processing electronics compare the differential transmissions of the two wavelengths and determine the species concentration.

The power requirements are 50 to 250 W for the transmitter, depending on the model used and the application. The receiver requires 60 W and can operate on 110 or 220 V ac. The transmitter weighs 17 kg, and the receiver weighs 18 kg. All optics are enclosed in sealed housings, and easy alignment is provided by pan tilt mirrors. The transmitted beam is 8 cm in diameter. Spacing between the transmitter and receiver can be varied from 5 m to 100 m, depending on the particular application. Typical applications are for continuous, unattended monitoring of potential hydrocarbon leak sources. Examples of monitoring purposes are:

- Pump rows
- Heat exchangers

- Propane units
- Pipe stills
- Loading areas
- Facilities for separating hydrocarbons from runoff water at refineries and tank farms
- Pipelines
- Tank farm perimeters.

The systems are capable of unattended operation and, under normal conditions, require maintenance only every 12 months. The systems can be operated in hostile environments. Although these instruments are designed for operation in hostile, industrial environments, hydrofluoric acid, hydrogen sulfide, and sulfuric acid vapors may damage the instruments by etching their optical components and corroding their electronic components. They normally operate at temperatures from -20 to 43 °C, although wider temperature ranges are available on a custom basis. An alarm system can be set at specified levels as determined by the application. A separate alarm is sounded when the system detects a transmission blockage for more than 30 s. Cost figures were not provided by the manufacturer. A number of these monitors have been sold to the chemical industry and have been used there for periods of up to 5 years. Specifications of the model DVX infrared hydrocarbon vapor monitor are included in the appendix, along with a partial list of species that have been detected with this system.

Computer Genetics Corporation (CGC) also offers a nonlaser optical system called SentiNet that can be used as a gas leak detector in a variety of scenarios. The system offered commercially as SentiNet is based on a series of continuous infrared emitters and receivers that are spaced along a pipeline or around a facility perimeter. Spacing of the emitter-receiver pairs can be varied from 10 m to 100 m. All emitter-receiver pairs are connected to a central controller that can trigger an alarm, display information on a video display, or print a hard copy record.

Detector response can be selectively tuned to provide measurement capabilities for a variety of industrial and hazardous compounds. A complete list of compounds that have been monitored by this system is included with a list of the system specifications in the appendix. The sensitivity varies for each compound and depends on the path length.

The system can operate on 110 or 240 V ac, 50/60 Hz. It is weather resistant and operates unattended in a continuous mode. Self-checking features have been designed into the system.

Cost estimates were provided by CGC in the form of a comparison to costs for fixed-point monitors to protect a hypothetical length of pipeline or perimeter with 100 percent confidence for a 5-year period. The cost of SentiNet emitter-receiver pairs for the detection of ethylene is $10,000 each. With installation, calibration, and maintenance costs, the SentiNet emitter-receiver pairs cost $17,250 each over a 5-year period. Each emitter-receiver pair would provide 100 percent coverage over a length of 100 m for a 5-year period. A list of specifications for this instrument is given in the appendix.

ELS also offers a double-ended long-path infrared system capable of detecting up to six gases simultaneously over a distance of more than 100 m. The system is similar to others using separate transmitter and receiver housings described in this report. Gases that have been monitored with the infrared system include methane, ethane, hydrogen cyanide, sulfur dioxide, hydrogen sulfide, hydrogen fluoride, nitric oxide, and hydrocarbon. The cost of the system is $30,000 to $40,000 for a transmitter-receiver pair, depending on the particular configuration.

Another long-path infrared absorption system using tunable diode lasers has been developed at McMaster University in Hamilton, Ontario, Canada.[77] Originally, this system was configured for open-path monitoring using a retroreflector that was separated from the laser and its detector by distances of up to 600 m. It was found that this configuration was subject to atmospheric turbulence. In addition, the true zero level could not be determined because it is impossible to remove the ambient air sample from the beam path. Consequently, the system was reconfigured by replacing the retroreflector with a multi-traversal White cell having a base path of 5 m. The cell allowed the path length to be varied quickly between 40 and 240 m. The problems associated with turbulence and zero-level determination were eliminated. The system could then detect isolated absorption lines in the atmosphere with a sensitivity of approximately 10^{-6} m^{-1} for a measurement time of a few seconds. This sensitivity allows nitrous oxide to be detected at a concentration of 20 ppb, methane at 2 ppb, carbon monoxide at 0.25 ppb, and ammonia at 0.75 ppb.

Other Canadian researchers at York University and Unisearch Associates have developed a similar tunable diode laser system for measurement of parts per billion levels of nitric oxide, nitrogen dioxide, and nitric acid in ambient air.[78] As above, a White cell was used to obtain long path lengths (i.e., 40 m) through a relatively small volume of captive ambient air. Minimum detectable concentrations for nitric oxide, nitrogen dioxide, and nitric acid in ambient laboratory air were found to be less than 0.5 ppb. This system has been used in comparison studies with a chemiluminescence system and in measuring tropospheric air in Los Angeles and southern Ontario.

Unisearch Associates of Concord, Ontario, Canada, have a tunable diode laser system that is mounted in a mobile laboratory and is available for hire. Their rates are approximately $40,000 to $50,000 for a 3-week program, although this will vary with traveling and living expenses. They have also sold eight of these tunable diode laser systems, largely for applications involving mobile laboratories. One sale has been to NASA for an airborne application.

DEVELOPING TECHNOLOGIES

Passive Techniques

Radiometers--

A device similar in principle to the Visiplume was developed by the U.S. Navy for infrared detection of clouds of nerve agents. Horman et al.[79] describe this approach, in which a forward-looking infrared (FLIR) thermal video system is used in combination with four selective infrared band-pass filters. One filter passes all incident-emitted radiation in the 8- to 14-μm range. A second filter minimizes the FLIR response to radiation emitted by the chemical agent. A third and fourth filter maximize the response to the two major infrared emission bands of agents. The four thermal images are then compared by determining the contrast variations of a suspect cloud relative to the background. Results of tests of the specific approach for military applications have not been made available. Various FLIRs for nonmilitary applications are available from several manufacturers, varying in cost from $30,000 to $2,500,000, depending upon spatial, temporal, and spectral resolution.

Discussions with optical engineers researching FLIRs for the military indicate much effort is currently in progress to miniaturize detectors, improve images, and reduce costs. Specifics of this research have been reported in Optical Engineering, Proceedings of the Society of Photo-Optical Instrumentation Engineers, and Proceedings of IRIS (Infrared Instrumentation Society). The last publication is classified.

This approach was also attempted in the 1960s by General Electric for the U.S. Army in a project called LOPAIR (Long-Path Infrared). This passive remote sensing instrument uses background sky radiation from 5 to 15 degrees above the horizon as its source.[80] The spectral region of interest is from 8 to 13 μm. This region has good atmospheric transmission and has relatively large amounts of natural radiant energy. The instrument consists of two subsystems: A spectroradiometer and a discriminator (or correlator). The spectroradiometer uses a circular, variable filter (rotating at 1 Hz), an optical chopper (rotating at 100 Hz) and a liquid-nitrogen-cooled, mercury-cadmium-telluride detector. The chopper alternately exposes the detector to background sky radiation and an internal reference blackbody. The interaction of the spectroradiometer's three components produces an ac signal that scans the spectral region in 0.01-μm increments. This signal is converted into a digital spec-trum by the discriminator, which is a small, special-purpose, hybrid computer using correlation techniques. Because of limits in the size of the computer, the digital spectrum has a resolution of 0.16 μm. By taking the difference between digital target and background spectra, the discriminator produces the infrared absorption spectrum of compounds of interest in the target. The instrument was tested using a small cloud of dimethylmethylphosphonate as the target gas. Optical depths of approximately 170 mg/m^2 were measured by the instrument in this test. Evaluations[81] found that absorption by ambient dust (seldom a problem at sea) in the 10-μm region was a major interferent.

Gas Correlation Spectrometers--

Prototype gas correlation spectrometers have been designed for various purposes, including temperature sounding of the atmosphere by monitoring emission of carbon dioxide;[26] remote sensing of several gaseous pollutants such as methane, ethane, hydrogen chloride, and carbon monoxide;[27] measuring the carbon monoxide levels in the troposphere;[72] and remote detection of methane leaks from natural gas pipelines.[71] The instruments are designed with either one or two detectors. In the one-detector system, background emission is continuous, but the emission from the target gas is chopped. The modulated radiation of the target gas is detected with a phase-sensitive detector. The two-detector systems use optoelectric gain balancing and stabilization. The gas correlation spectrometers that have been built so far have been limited to the measurement of a single-target gas species. However, it is theoretically possible for multiple species to be measured simultaneously. Detection limits for remote sensing include 50 ppm-m for methane and ethane, 60 ppm-m for carbon monoxide, and 300 ppm-m for hydrogen chloride.[27]

IRGIS--Developed for the Canadian military for chemical agent detection, the IRGIS (infrared gas imaging system) device incorporates an infrared video system as the detector in gas correlation spectrometry (C. Cumming, Barringer Research, Ltd., personal communication, 1987). The specificity of GASPEC is retained and enhanced with the spatial information obtained from video images. No price could be quoted for another system, which would require further development for application to other gases.

Selective Modulation Radiometer--This type of gas correlation spectrometer was described by Guillot et al.[70] It was evaluated in field measurements by comparing it with mask correlation spectrometers (COSPEC and TELETEC--commercial instruments, described above). Instead of one reference gas cell that is spectroscopically inert and one gas cell containing sample gas to be detected, this device uses four gas cells, each containing different amounts of the sample gas. The four gas cells are used in pairs, and each pair is illuminated alternately to produce a modulated signal, the difference between maxima and minima being proportional to the sulfur dioxide optical depth. It was susceptible to the same errors as the COSPEC because of spectral and temporal variations in the background skylight, and the same onsite calibration procedures as for COSPEC are followed. Apparently this device, developed by J. Laurent of Office National d'Etudes et Recherches Aerospatiales, Chatillon-sous-Bagneux, France, has not been discussed directly in the English literature, because a literature search based on that name did not locate further information. The expressed intention of the CEC, who supported its development, was that it would be commercialized.

Interferometers--

Honeywell, Inc., of Clearwater, Florida, has built a passive ROSE-like instrument called the XM21 Remote Sensing Chemical Agent Alarm.[82] This instrument is an automatic scanning, infrared sensor for detection of nerve agents and is currently being tested by the U.S. Army. The goal of this program is to provide a portable sensing device that can detect chemical agents at a level of 150 mg/m^2 at a range of 3 to 5 km. The sensor is a Fourier transform spectrometer operating in the 1,200 to 800 wavenumber region of the infrared spectrum. The system operates in an unattended mode for extended periods of time and is simple to set up. Major components of the system are a Michelson interferometer coupled with an onboard microcomputer. The interferometer yields the sensitivity required for operation under varied meteorological conditions while the microcomputer controls the system, analyzes the data, and makes agent/no-agent decisions. The entire system is packaged for operation in a battlefield environment and can be easily operated and maintained. The detector and its tripod weigh 29 kg. The system requires less than 80 W from an external thermoelectric generator and 1 to 4 psig pressurization for the interferometer's mirror bearing. Although this instrument was developed specifically for the measurement of nerve agents, its successful development indicates a potential for the remote sensing of other organic compounds with infrared emission bands using interferometric techniques.

Heterodyne Radiometry--

In recent years, progress has been made in the area of heterodyne detection of spectral lines in the infrared region; however, the technique is also promising for passive remote sensing. Recently, interest in heterodyne detection has increased because it offers high sensitivity, high spectral resolution, and high spatial directivity. These spectrometers can be used to monitor either thermal emission of atmospheric species or solar absorption by these species.[83,84]

In brief, optical heterodyne detection is a means to reduce the impact of noise on measurements in the middle infrared region (i.e., 1 to 10 μm wavelengths). For directly detected, backscattered signals in this region, noise due to background blackbody radiation is much greater than detector noise and the signal-to-noise ratio is low. However, such a signal may be mixed in a heterodyne detector with a more powerful signal (e.g., 0.2 mW vs. 1 pW) from a local oscillator laser. The frequencies of these two signals will differ by a few megahertz. The resulting heterodyne signal emerging from the detector will be at the difference frequency and can be processed by narrow bandwidth, radio frequency electronics. The heterodyne signal's strength is proportional to the strengths of the two input signals. It will be dominated by narrow-bandwidth, local oscillator noise that can be time averaged rather than by wide-bandwidth background noise. The signal-to-noise ratio for a heterodyne detector at a 10-μm wavelength may be a factor of up to 10,000 times greater than that for a direct detector (dependent on bandwidth and backscattered signal power). However, it should be noted that the complexity of a heterodyne detector is much greater than that of a direct detector because of the addition of the local oscillator laser and the associated electronics and optical components.[4-5,85-86]

Both gas lasers (most notably carbon dioxide) and tunable diode lasers (TDLs) have been employed as local oscillators in these instruments; however, present technology of TDL does not meet the required power, noise, and mode purity for highly sensitive heterodyne detection.[85] To date, the technique has been used mainly to gather astrophysical data, but it has also been used in the atmospheric detection of several species, including carbon dioxide, ozone, nitrous oxide, sulfur dioxide, ammonia, and methane.[87-91] The sensitivities for many of the species are in the low-ppb range.[91] Because it has better spectral resolution than any of the other passive remote sensing techniques discussed, heterodyne radiometry is the passive technique that is most suited to measuring the concentration of target gas at long distances from the detector. Near a wavelength of 10 μm, which is the common region for heterodyne radiometers, the resolution is good enough to obtain profiles up to 35 km.[92]

A comparison of the heterodyne radiometer with FTIR is of interest. Multiplexing with Fourier transform spectroscopy produces large increases in the signal-to-noise ratio until the fluctuations of background radiation become the predominant noise source.[92] This limitation becomes more severe toward longer wavelengths. The heterodyne technique, however, reduces both the detector noise and background radiation noise, but requires operating with large bandwidths and narrow fields of view. The heterodyne radiometer also interacts with one spatial mode, which results in a wavelength-dependent field of view. The heterodyne radiometer becomes more advantageous at longer wavelengths (>8 μm) and in spectra containing fewer resolution elements.[93] Heterodyne radiometry is also limited by local oscillators. Only selected spectral intervals where an appropriate local oscillator exists can be measured by this technique. The FTIR interferometer has the advantage of simultaneously obtaining a measurement over the entire region from 5 to 14 μm.

Active Techniques

Long-Path Absorption--

A long-path measurement system was operated in a van as a joint venture by Lincoln Laboratory and U.S. EPA.[94-96] It used a tunable diode laser for multipollutant detection and a lens-mirror retroflector as the target. Although this system was used primarily for ambient monitoring, it could be applied to source monitoring for nitric oxide, nitrogen dioxide, sulfur dioxide, and carbon dioxide.[94]

The U.S. EPA has compared this tunable diode laser system with a long-path measurement system using a carbon dioxide laser.[96] The latter system was developed at the General Electric Electronics Laboratory. It also used a retroreflector to return light to its detector. The carbon dioxide laser produces light having a number of fixed and closely spaced wavelengths. This laser differs from the tunable diode laser that produces light having a variable wavelength. The carbon dioxide laser system measured ozone and showed good agreement with point ozone monitors. Its closely spaced wavelengths minimized system drift and simplified the determination of ambient ozone concentrations.

A long-path carbon dioxide laser system is used in Sweden for application to source monitoring.[97] The application discussed is the measurement of ethylene mass flow from a petrochemical factory. The carbon dioxide laser was computer-controlled and operated with a series of reflectors located on a vertical mast. By sequentially targeting the separate mirrors, the laser provided a cross-section of ethylene concentration in the region of interest. Combined with windspeed data, the ethylene concentration determinations allowed an assessment of ethylene mass flow from the facility.

This same computer-automated, carbon dioxide-laser long-path absorption system has been used for mass flow estimations and plume profile measurements of diffuse ethylene leakages from a petrochemical plant.[98,99] Where the laser previously had been aimed at a number of retroreflectors set vertically on a mast, it was now aimed at a number of retroreflectors set at various positions throughout the plant's area. A computer controlled the aiming of the laser via a motor-driven mirror. Sulfur hexafluoride was released as a tracer gas and was also measured by the long-path absorption system. The ethylene mass flow was calculated by measuring the concentration levels of ethylene and sulfur hexafluoride leaving the factory area and by using the known release rate of the tracer gas. In situ measurements of ethylene and sulfur hexafluoride were also performed. The average concentrations obtained from the in situ measurements were in very good agreement with those obtained from the long-path absorption system.

Differential Optical Absorption Spectroscopy (DOAS) should be mentioned as a subset of long-path absorption spectroscopy that holds some promise for source-oriented monitoring applications. The basic concept of this technique is the same as for long-path absorption spectroscopy except that multiple wavelengths are measured. One wavelength is located at the center of a strong absorbance line of a species of interest. The other wavelength is nearby, but not on the absorbance line. The concentration of the species of interest is determined by the difference in the absorbance at the two wavelengths. Interferents such as water vapor and carbon dioxide are not detected because they have broad spectral features that absorb nearly equally at both wavelengths.

The technique was developed in the Federal Republic of Germany and was applied to the measurement of nitrous acid in the atmosphere.[100] In this application, a powerful broadband light source was mounted in a reflector arrangement of a distance of a few kilometers from an optical receiver system operating in the ultraviolet and visible spectral region. A repetitive fast scanning over a small spectral region was made by a dispersive spectrometer. The rapid scan rate left even minute atmospheric absorption features unaffected by atmospheric turbulence, which otherwise frequently limits atmospheric long-path optical measurements. For a pathlength of 5 km and 20 m monitoring time, the technique had a background noise level equivalent to

24 ppt nitrous acid. Concentrations as high as 0.8 ppb were measured in moderately polluted air.

DOAS has also been applied to measurements of atmosphere mercury, sulfur dioxide, and nitrogen dioxide in Sweden.[101] This system uses a high-pressure xenon arc lamp as the light source. A concave mirror collimates the light and directs the resulting beam to a telescope that is located between 0.1 and 10 km away. A fast-scanning dispersive spectrometer and a photomultiplier tube detect the light captured by the telescope. For a pathlength of 0.6 km, the system has a background noise level equivalent to approximately 4 ppb for nitrogen dioxide. Mercury concentrations on the order of 2 ppt were measured over a 0.7-km path. It is understood that this sytem is under commercial development by Opsis AB of Lund, Sweden.

No source-monitoring applications of DOAS systems have been located in the open literature. However, it is feasible that such applications will be forthcoming.

Differential Absorption Lidar (DIAL)--

Differential absorption lidar is more complex than long-path monitoring. DIAL systems are more common, however, even for source monitoring applications, because they offer improved specificity and ranging capabilities, and are generally open-ended (i.e., do not need a cooperative target). However, infrared DIAL systems do not have good range-resolved capabilities and so may need cooperative targets of known characteristics (J. L. McElroy, U.S. EPA, Las Vegas, personal communication, 1988).

Computer-controlled DIAL systems can be used to scan a volume to provide a three-dimensional map of concentration. This capability makes them particularly useful for scanning the emissions from a large facility that has several point and area sources. Using DIAL, the entire facility can be scanned from one location every few minutes.

A tunable dye laser system has been used to monitor nitrogen dioxide concentrations over a chemical factory in Germany.[102] The same researchers have also used a carbon dioxide laser system to monitor ethylene concentrations around a refinery.[103] The same dye laser system was used to monitor nitrogen dioxide concentrations in ambient air downwind of a city.[104] Because of the large signal return from the source monitor application, an analog detector was used instead of the photon counter used for ambient applications. Concentrations were measured at a distance of 300 m from the stack. An additional experiment was conducted to survey the entire plant area. Within minutes, the distribution of nitrogen dioxide over the plant site could be mapped.

A mobile lidar system for environmental probing was developed at the Chalmers University of Technology and the Lund Institute of Technology in Sweden.[105-108] The system is operated primarily in the DIAL-mode. However, it can also be operated as an atmospheric backscatter lidar, a Raman backscatter lidar, and a DOAS system. The light source for the system is a dye laser pumped by an Nd:YAG laser in the first three modes. An external light source is needed for operation in the DOAS-mode. The system has been used for ambient air quality and source emissions measurements of nitrogen dioxide and sulfur dioxide in the DIAL mode. For example, nitrogen dioxide was measured in the plumes of nitric acid and saltpeter plants at distances of between 300 and 400 m from the plumes. It was also measured in city streets in the urban plume from a major city. Sulfur dioxide was measured in the vicinity of a paper mill and in the distributed plume downwind from a hydrochloric acid factory. The authors also describe modifications that could be made to permit dual-beam lidar measurements of plume velocity using this system.

Recently, the group at the Lund Institute of Technology has constructed an improved version of their mobile DIAL system for atmospheric monitoring.[109] While it is at present arranged for DIAL measurements, the new system is intended for all kinds of exploratory optical remote sensing work (e.g., gas correlation lidar, fluorescence lidar, Raman lidar, and DOAS). The light source is a dye laser pumped by an Nd:YAG laser with options for frequency-doubling and Raman-shifting. In general, the optical components are improved over those of the earlier DIAL system. This DIAL system has been used for sulfur dioxide and aerosol measurements in the plume from a paper mill. The system was approximately 1 km from the plume. For ranges up to 1 km, the detection limit for the integrated sulfur dioxide concentration is 2 ppm-m. The DIAL system has also been used for nitrogen dioxide measurements in a ground inversion situation.

A number of ground-based and airborne DIAL systems have been developed at NASA's Langley Research Center (NASA/Langley) for measurements of tropospheric gases.[110] One of these ground-based systems was used for investigations of sulfur dioxide and aerosols in power plant plumes. In this system, a double-pulse, frequency-doubled Nd:YAG laser is used to sequentially pump two independently tuned dye lasers. The two pulses are separated in time by 100 μs. This close temporal spacing minimizes concentration errors resulting from changes in atmospheric scattering characteristics during a DIAL measurement. The typical ultraviolet light output is 2.5 mJ. Sulfur dioxide measurements were made in the plume of a Maryland power plant during the night. Horizontal scans were made across the plume 275 m above the stack exit with the lidar system 1.3 km from the measurement plane. The path distance through the plume at the center was 274 m. The average sulfur dioxide concentration across the plume was then 1,300 ppm-m/274 m, or 4.7 ppm. This value is within 18 percent of the expected concentration at that plume diameter from in-stack measurements and within 5 percent of the predicted value from the sulfur content in the fuel mixture. It was concluded that an ultraviolet DIAL system can obtain the extent and average sulfur dioxide concentration in power plant plumes and that, with increased ultraviolet laser power, range-resolved sulfur dioxide measurements can be made with an airborne DIAL system at long ranges downwind of the stack. The NASA airborne DIAL system has the capability to make such measurements.

Two ground-based DIAL systems were used in a French study of the dispersion of sulfur dioxide and ozone in the planetary boundary layer.[111] Both systems used dye lasers pumped by Nd:YAG lasers and second harmonic generation crystals to produce light in the spectral region from 280 to 320 nm. One system measured range-resolved ozone concentrations over a 3-km pathlength for most of the time during the study. Good agreement was found between these measurements and those of an in situ monitor. The second system was used for sulfur dioxide measurements in the planetary boundary layer and downwind of various point sources. Vertical concentration profiles were obtained at distances up to 2.5 km from the system and at altitudes up to 0.9 km above it. The system's measurements showed good agreement with those from Italian and English mobile lidar systems during a brief intercomparison study. Both French systems had a spatial resolution of 500 m, a temporal resolution of 3 min, and a maximum horizontal range of 3 km. The overall accuracy as assessed by the various method intercomparisons was always better than 20 percent.

A group at the Massachusetts Institute of Technology has developed a DIAL system for remote sensing of ammonia in the atmosphere.[112] A dual carbon dioxide laser is the light source. For backscattered return from topographic targets at ranges up to 2.7 km, the path-averaged ammonia sensitivity was 5 ppb.

One of the primary centers for atmospheric remote sensing research is SRI International, Inc., in Menlo Park, California. Although SRI is principally involved in aircraft-mounted DIAL lidars for regional scale applications, it has also developed some source measurement applications.

A system was developed by SRI for the Gas Research Institute (GRI) for the detection of ethane.[113] The application of interest to GRI was to detect gas leaks from natural gas pipelines. Ethane was selected as a target species because it is present in percent concentrations in commercial natural gas and would not be subject to false indications from natural methane sources.

No appropriate laser was available that emits at an ethane absorption wavelength convenient for DIAL measurements. The solution was to frequency-mix radiation from two carbon dioxide lasers that emit in the 9- to 11-μm range to produce radiation of the desired wavelength range at 3.4 μm.

A laboratory version of the system was tested and shown to be as effective for the detection of gas pipeline leaks as the manual approach now in common use. The system was located 46 m from the simulated test leak. The ultimate application of the system for the gas industry is to mount it on a van so that gas distribution lines can be monitored routinely at urban traffic speeds.

A ground-based DIAL system was developed by SRI to measure the atmospheric distribution of several pollutants (i.e., sulfur dioxide, ozone, and nitrogen dioxide) that are commonly associated with fossil fuel combustion.[114] This sytem also uses Nd:YAG laser-pumped dye lasers and frequency-doubling crystals as its light source. The system is mounted in a 40-ft long highway van for mobile applications. It has been used to measure plume and ground-level sulfur dioxide concentrations at 1 to 3 km downwind of a coal-burning power plant. These measurements showed good agreement with an in situ monitor's data. The system has also measured vertical ozone profiles in an urban area on a smoggy day. Good agreement was found between its measurements and those of an airborne in situ monitor. The system has also been used in a number of other source-oriented applications.

SRI has used an airborne DIAL system to map cross-plume vertical distributions resulting from a near-surface sulfur hexafluoride tracer gas release.[115] This system used a carbon dioxide laser for measurements in the 9 to 11 μm spectral region. In a number of demonstration tests, the system overflew an area of rolling terrain after the point release of the tracer gas. The system measured the column content and vertical concentration distribution of the tracer gas for 20 to 30 min after the release. The detection capability for the system was approximately 2 ppb-km for the column-content measurements and approximately 20 ppb for the altitude-resolved measurements. The tracer gas release and measurement technique may be useful for studying the accidental release of hazardous materials and for validating atmospheric dispersion models.

A mobile atmospheric pollutant mapping system is being developed at the California Institute of Technology's Jet Propulsion Laboratory for long-distance, range-resolved measurements of organic solvent vapors and aerosol clouds.[116,117] A pair of pulsed carbon dioxide lasers are used as the light sources and a pair of continuous-wave carbon dioxide lasers are used as local oscillators for heterodyne detection. This DIAL system is mounted in a 35-ft transportable semitrailer. It has been used to measure water vapor concentrations in the atmosphere. The horizontal measurement range was up to 6 km with a range resolution of approximately 200 m. The DIAL system measurements showed good agreement with psychorometer measurements. The system has been modified to increase the range to 8 km and to improve the accuracy.

Raman Lidars--

Optech, Incorporated, of Downsview, Ontario, Canada, is developing a remote sensor for natural gas leak detection for the British Gas Corporation.[118] The system in development relies on Raman scattering as the detection principle. The laser is a xenon chloride excimer laser, emitting at 308 nm. The pulse width is 4 ns and produces 3 to 5 W for operation at 100 Hz. It is controlled by a PDP 11 computer. It uses a 0.5-m-diameter Cassegrain reflecting telescope and has two detection channels, one for nitrogen and one for methane.

The system selectively detects methane concentrations of 2 to 20 percent at a distance of 100 m to 1 km with accuracy of 10 percent. It is carried on a truck, and power is supplied by an onboard generator. It occupies a space of 2.5 m x 6.1 m.

Gas Imaging--

BAGI is a technology that produces a visible image of an invisible gas cloud or plume.[119] Currently, BAGI technology is limited to qualitative location and real-time imaging of gas clouds. Quantitative measurements of gas concentration are not possible.

The principle of the technique is to direct a laser source into the area to be monitored. If the gas is present, the appropriate wavelength will be absorbed in the gas cloud but not outside the gas cloud. The amount of backscatter radiation at the absorption wavelength indicates the presence or absence of the gas cloud. The technique relies on differential absorption at two locations to sense the presence of the cloud. Video camera equipment and electronic signal-processing equipment convert the information into a video image that can be displayed on a standard television monitor.

The 20- to 25-W carbon dioxide laser operates at its natural beam divergence. The laser beam must be scanned in synchronization with the television raster scan. Table 2 lists species that can be detected by the BAGI technique. The system weighs approximately 64 kg, exclusive of the television monitor. Estimated cost is approximately $60,000 per unit.

POTENTIAL DEVELOPMENTS

Technology advances in laser research, video imaging, light detection, electronics systems for signal processing, and fiber optics will undoubtedly be applied to new atmospheric monitoring systems. However, manufacturers are hesitant to discuss systems that are in the development stage.

Research is now proceeding on multiple frequency lidars that will be capable of detecting more than one species or more than one particle size simultaneously. As part of SRI research for GRI, work is proceeding on a hand-held laser system for monitoring gas leaks. Details of the system were not available. The BAGI technology is proceeding, and future plans include mounting systems on aircraft, improving sensitivities, and developing long-range, multiwavelength systems.

TABLE 2. CURRENT BAGI CO_2 LASER GAS ABSORPTION DATA BASE AND DETECTION SENSITIVITY ESTIMATES

Gas	TLV-TWA (ppm)	Laser wavelength (μm)	BAGI sensitivity (ppm)
Acetonitrile	40	9.29379	200
Acrylonitrile	2	10.30347	17.2
Allyl alcohol	?	9.69483	13.8
Ammonia	50	10.34928	2.5
Benzene	10	9.62122	41.5
Butadiene	1,000	9.22953	2.4
t-Butanol	100	10.74112	21.6
Carbonyl difluoride	?	10.61139	15.2
Chloroprene	25	10.67459	30.2
Cyclohexane	300	9.62122	200
Cyclopentane	?	10.74112	876
1,2-Dichloroethane	100	10.59104	160
Dimethylamine	10	9.75326	97
Ethylacetate	400	9.45805	6.7
Ethylene	5,500	10.53209	2.9
Ethylmercaptan	0.5	10.19458	146
Freon-12	1,000	10.76406	1.7
Freon-113	1,000	9.60357	4.1
Furan	?	10.18231	19.9
n-Hexane	50	9.34176	441
Hydrazine	0.1	10.44059	11
Isopropanol	400	10.49449	21.9
Methacrylonitrile	1	10.61139	37.9
Methanol	200	9.67597	3.8
Methyl bromide	5	10.61139	122
Methyl chloride	?	9.60357	204
Methyl chloroform	350	9.24995	8.4
Methyl ethyl ketone	200	10.59104	68.6
Methyl methacrylate	100	10.61139	17.6
Monochloroethane	?	10.27445	25.2
Monomethylhydrazine	0.2	10.33370	24
Orthodichlorobenzene	50	9.62122	10.8
Ozone	0.1	9.50394	6.6
Pentane	600	9.67597	848
Perchloroethylene	100	10.74112	17
Propane	1,000	10.81111	5.8
Sulfur dioxide	5	9.21969	758
Toluene	100	9.62122	124.4
Trichloroethylene	100	10.59104	6.6
Trimethylamine	10	9.58623	20.1
Unsymmetrical dimethylhydrazine	0.2	10.83524	21.1
Vinyl chloride	5	10.61139	9.5
Xylene	100	9.53597	95.8

TLV = Threshold limit value. TWA = Time-weighted average.
BAGI = Backscatter/absorption gas imaging.

SECTION 4

PRACTICAL CONSIDERATIONS

GENERAL

Remote sensing technology is highly applicable to atmospheric monitoring. The ability of remote sensors to sample large volumes of air rapidly provides significant advantages over traditional point sensors for air pollution measurement applications. The advantages offered through remote sensing are particularly applicable to regional-type monitoring from aircraft platforms.

Another application of remote sensing that is receiving considerable attention is space-based system development. Systems are in development that will measure windspeeds and directions, temperature profiles, and atmospheric pressure, as well as pollutant species. Often, the costs of developing and operating these complex remote measurement systems are great. The advantages of applications to air pollution source emissions measurements sometimes do not justify the costs associated with commercial development. This is particularly true in the application of active, ranging, remote sensors to fixed-point source emission monitoring.

Many applications of remote sensing technologies to source emissions monitoring have been discovered, and several have become commercial ventures. In general, passive systems have been applied to routine monitoring of source emissions such as smokestack-type emissions, and active techniques have been applied more often to area or line sources involving potential accidental releases of explosive or hazardous materials. The companies that offer commercial pulsed laser measurement systems also offer van-mounted systems that can be leased at a cost considerably less than the purchase price.

A significant number of pollutant species can be detected by remote sensing, and that number will increase with continuing research. Technologies exist that can be used to measure almost any air pollutant by remote sensing principles. But the investment of labor and money to develop, build, and test a system can often be significant and even prohibitive. Thus, every application should be considered individually to determine if benefits justify the potential costs.

ADVANTAGES AND LIMITATIONS

Passive Systems

There are limitations to passive remote measurement systems that rely on natural radiation. One is the dependence on favorable meteorological conditions. Light source intensities vary widely in different locations, during different seasons, and during periods of the same day at the same location. Compensation for these variations often requires additional steps in the signal processing and data reduction, and short-term variations still contribute to error. When the sun is not in direct view of the system, light sources are weak, and pulsing or chopping of radiation from the source is generally not practical, so extraneous radiation becomes more of a problem than with active techniques. This leads to an unknown zero offset common to many passive techniques. The significance of this error varies widely from situation to situation. For infrared measurements, the atmospheric conditions are also important because they determine the effective temperature difference between the target emissions and the background black-body radiation that determines the signal-to-noise ratio. Another limitation of passive remote sensors is the inability to determine the amount of emissions as a function of distance from the detector. Thus, range-resolved spatial mapping of the gas usually cannot be obtained. Rather, the measurement is the integral concentration of emissions along the entire line of sight.[120] The quantity measured is optical depth, the product of a concentration and a pathlength.

These limitations do not preclude the use of passive devices for remote monitoring of source emissions. Passive sensors are generally simpler, more convenient, easier to operate, and easier to maintain than active sensors. They are also usually considerably less expensive than active sensors. In one comparison of an active and passive monitor for sulfur dioxide, the performance of the passive sensor (i.e., a correlation spectrometer) was equal to that of the active sensor (i.e., DIAL) in accuracy and sensitivity.[120] A physical and economic evaluation of the two systems was also included in the referenced report. The active system weighed 25 times as much and cost 10 times as much as the passive system. Thus, passive monitors are often capable of providing the desired information more cheaply than active remote sensing systems.

Active Systems

Active systems have severe limitations as well. They are often cumbersome, they sometimes require special power supplies, and optics alignment can require regular maintenance and attention. Pulsed laser systems, used to obtain range-resolved measurements, require sophisticated signal-processing electronics and significant data storage capacity. The operation of the system, data handling tasks, data interpretation, and routine system maintenance often require a skilled technician trained for the task.

Advantages of active techniques are related to extent of coverage either spatially or temporally. The spatial coverage offered by either computer-controlled or mobile, mounted systems is obvious. A sweep of an entire chemical complex can be accomplished in several minutes by using a computer-controlled laser-based system. A mobile system mounted on a truck or a van can define the extent and characteristics of a widely dispersed power plant plume in a short time.

The temporal coverage provided by continuous long path systems makes them well-suited for perimeter monitoring and pipeline surveillance in cases where hazardous materials can be released inadvertently. Long-lasting light sources and simple electronics that focus on alarm level detection make these systems capable of continuous unattended operation for long periods of time. Often, lasers are not required or even desired for this type of application, and the simplicity that can be tolerated brings the cost down to levels competitive with passive remote sensing systems.

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APPENDIX

Specification lists and lists of species that can be measured by several of the commercially available instruments described in this report are presented here. The instruments included are as follows:

- Barringer Research COSPEC^{TM} V Correlation Spectrometer
- Barringer Research GASPEC II Non-Dispersive Gas Filter Spectrometer
- Barringer Research Mini-COSPEC Correlation Spectrometer
- Computer Genetics Corporation Mobile Particulate Lidar
- Computer Genetics Corporation SentiNet Long-Path Infrared Hydrocarbon Monitor
- Environmental Laser Systems LASERSAFE DIAL Lidar
- Recon Explorations GASPILS Gas Correlation Spectrometer
- Recon Explorations Plumetracker Differential Absorption Spectrometer
- Research Ventures Visiplume Ultraviolet Video-Radiometer, and
- Wright and Wright Model DVX Open-Path Infrared Hydrocarbon Monitor.

Contacts for the above companies are:

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Barringer Research Ltd.
304 Carlingview Drive
Rexdale, Ontario
Canada M9W 5G2
(416) 675-3870

Mark A. Maris
Computer Genetics Corporation
18 Lakeside Office Park
Wakefield, Massachusetts 01880
(617) 246-2838

Orman A. Simpson
Environmental Laser Systems, Inc.
3333 Chamblee Dunwoody Road
Atlanta, Georgia ,30341
(404) 457-0977

C. Keith Thompson
Recon Exploration
15506 Wright Brothers Drive
Dallas, Texas 75244
(214) 960-8600

Reginald J. Exton
Research Ventures, Inc.
P.O. Box 1280
Williamsburg, Virginia 23185
(804) 220-3430

Linda J. Stahl
Wright and Wright, Inc.
P.O. Box 1728
Oak Bluffs, Massachusetts 02557
(617) 693-2608

BARRINGER RESEARCH COSPEC™ V SPECIFICATIONS

Target gases	Sulfur dioxide or nitrogen dioxide
Light source	Skylight
Operational modes	Vertical look (plume diffusion), side look (plume rise); tripod, or mounted in ground mobile vehicle or in aircraft
Sensitivity at a peak-to-peak, signal-to-noise ratio = 1	*Sulfur dioxide = 2.5 ppm-m; nitrogen dioxide = 2.5 ppm-m
Dynamic range	*Normally 1 to 1,000 ppm-m larger ranges can be calibrated on request
Baseline temperature stability	*1 ppm-m/°C
Operating temperature range	*0 to 50 °C
Diurnal baseline drift	*Better than 40 ppm-m from sunrise to sunset
Response time (integration time)	1 to 32 s in 5 switch-selectable steps
Output	Nominally: 0 to 10 V; Signal output impedance - 10 kΩ AGC output impedance - 220 Ω
Calibration	Two fused silica cells of known sulfur dioxide (or nitrogen dioxide) burden, internal reference quartz-iodine light, micrometer screw grating adjuster with guard
Optical system	Direct reading Cassegrain telescope
Field of view	10 x 30 mrad
Spectrometer	1/4 meter F/5.6 Ebert-Fastie
Power requirements--general operation	12 V dc: 115/230 V ac: 8:23 W or 24 V dc: 115/230 V ac: 11:23 W
Weights and dimensions	
--Instrument	19.3 kg; 78.1 x 37.5 x 22.2 cm
--Carrying case	20.5 kg; 101.6 x 53.3 x 28 cm

*Collected under laboratory conditions.

BARRINGER RESEARCH GASPEC II SPECIFICATIONS

Specifications	
Target gases	To be selected from table of target gases or upon request. Barringer Research, Ltd. (BRL) will make a preliminary evaluation of any gas of customer interest. An optional dual gas configuration is possible for species with conveniently located spectral absorption bands.
Light sources	Ambient infrared emission, scattered solar radiation, or active artificial source.
Optics	Field-defining telescope may be chosen to suit. Standard unit has a 20-mrad FOV lens.
Dynamic range	Gas dependent. Typically in excess of two orders of magnitude gas burden.
Response time	Typically fixed at 1 s, can be modified to suit customer requirements.
Outputs	Gas signal: 0 to 10 V dc at 10k Ω output resistance Radiometer output: 0 to 10 V dc at 10k Ω output resistance Other outputs: selected test points for monitoring instrument operation.
Controls	Power ON/OFF Zero adjust Time constant (optional) LED dynamic range indicators Power source selector (optional) Calibration cell selector (optional) Test points
Mechanical	Size--13 in. x 10 in. x 26 in. (33 cm x 25 cm x 63 cm) approximate Weight--40 lb (18 kg) approximate Mounting--mounting plate provided with standard tripod mounting holes.
Environmental	Ambient temperature range: 0 to 50 °C
Power requirements	80 W, 115 V, 60 Hz (can be configured for dc operation on request)
Options	A wide range of options available to suit individual requirements.

BARRINGER RESEARCH GASPEC II
POTENTIAL TARGET GASES AND TYPICAL SENSITIVITIES

Species	Wavelength, μm	Estimated detection limit, ppm-m (at 300k)
Carbon monoxide	4.6	10
Carbon dioxide	4.3	20
	15.0	30
Methane	7.66	100
	3.31	1,000
Ethane	12.0	200
	3.4	400
Hydrogen sulfide	7.8	1,000
Sulfur dioxide	8.6	50
	7.3	25
	4.0	3,000
Nitric oxide	5.3	50
Nitrous oxide	3.45	4,000
	17.0	200
	7.78	20
	4.5	200
Ammonia	11.0	10
	6.15	100
Hydrogen chloride	3.6	450
Hydrogen bromide	3.9	400
Hydrogen cyanide	14.0	20
	3.2	500
Carbon tetrafluoride	15.8	11
	7.9	200
Carbon disulfide	6.55	22
Silicon tetrafluoride	9.7	1
Sulfur hexafluoride	10.6	1
Acetylene	13.5	15
	7.5	30
	30.4	1,000
Ethylene	3.34	5,000
	3.12	3,000
Trichlorofluoromethane	11.9	5

Other gases include: Acrolein, aminomethane, benzene, carbon tetrachloride, freon-12, furfural, naphthalene, nitric acid, propane PNAs (polynuclear aromatic hydrocarbons), and vinyl chloride.

BARRINGER RESEARCH MINI-COSPEC^{TM} SPECIFICATIONS

Target gases	Sulfur dioxide or nitrogen dioxide
Light source	Skylight
Operational modes	Vertical look recommended; tripod, or mounted in ground mobile vehicle or in aircraft
Sensitivity at a peak-to-peak, signal-to-noise ratio = 1	Sulfur dioxide = 4 ppm-m; nitrogen dioxide = 4 ppm-m
Dynamic range	Normally 1-1,000 ppm-m larger ranges can be calibrated on request
Baseline temperature stability	1 ppm-m/°C
Operating temperature range	0 to 50 °C
Diurnal baseline drift	Better than 50 ppm-m from sunrise to sunset
Response time (integration time)	1 to 32 s in 5 switch-selectable steps.
Output	Nominally: 0 to 10 V; Signal output impedance - 10 kΩ AGC output impedance - 220 Ω
Calibration	Two fused silica cells of known sulfur dioxide (or nitrogen dioxide) burden, screw grating adjuster
Optical system	Direct looking
Field of view	10 degrees x 10 degrees
Spectrometer	1/4 meter F/5.6 Ebert-Fastie
Power requirements--general operations	12 V dc: 8 W
Weights and dimensions	
--Instrument	15 kg; 65 x 37.5 x 18 cm
--Carrying case	16 kg; 86 x 44.4 x 25 cm

COMPUTER GENETICS CORPORATION MOBILE PARTICULATE LIDAR SPECIFICATIONS

Receiver/Transmitter

Wavelength: Tunable dye (350 to 750 nm)
Beam diameter: 6 mm
Beam divergence: 1.5 mrad
Maximum power: 4.5×10^6 W
Pulse width: 10 to 20 ns (selectable)
Coaxial telescope (12 in.)
3 Primary separate transmitters
RCA 8850 or 7256 photomultiplier
1-nm bandpass filters (6 max.)

Data Acquisition and Control Electronics

Data General central processing unit s/64 kb, Cos System Biomation 8100 (2048, 8 bits, 10 ns), Biomation 6500 (2048, 6 bits, 2 ns) or LeCroy 2249SG charge digitizer (10 bits, 10 ns)

Output Devices

Texas Instruments, 810 bi-directional printer or ASR-33 teletypewriter
Infoton 100 CRT display
9-track industry compatible magnetic tape

Scanner Drive

Manually controlled azimuth axis (360 degrees)
Motor-controlled elevation axis (horizon = 20 degrees + 200 degrees)
Computer drive stepping motor, 0.05 degrees elevation resolution

Vehicle

1979 Dodge Kary Van
6.5 kW Onan generator

COMPUTER GENETICS CORPORATION SENTINET™ LONG-PATH INFRARED SYSTEM SPECIFICATIONS

Environmental Operating Range

Temperature: -10 to +130 °F (-23 to +54 °C)
Humidity: 0 to 90 percent, noncondensing

Sensitivity

50 to 100 ppm averaged over nominal 100 m of concentration path length.

Sensor Spacing

100 m nominal interval (actual interval determined by user's requirements)

Monitoring Mode

Continuous

Configuration

Remote sensing

Power Requirement

110/240 V ac, 50/60 Hz

Light Source

Invisible, infrared light source, long-life type

Construction

All solid-state circuitry; weather-resistant, industrial-grade cylindrical aluminum housing

Optional Equipment

Color monitor, dot-matrix printer, audible/visual alarm

PARTIAL LIST OF COMPOUNDS DETECTABLE BY COMPUTER GENETICS CORPORATION SENTINET^{TM}

Chlorine
Acetic acid
Acetylene
Hydrogen cyanide
Isoprene
Methane
Ethylene
Formaldehyde
Methyl mercaptan
Ammonia
Naphthalene
Acetone
Methylene chloride
Nitrogen dioxide
Acrylonitrile
Phenol
Ethyl alcohol
Carbon monoxide

Sensitivities vary for each compound, depending on optical path length. Examples of detection thresholds are 0.016 ppm for chlorine (sensors spaced at 10 m) to 100 ppm for nitrogen dioxide (sensors spaced at 150 m). These thresholds may be varied greatly by changing the spacing of SentiNet units. Contact Computer Genetics Corporation for information regarding sensitivities for specific compounds and sensor geometries.

ENVIRONMENTAL LASER SYSTEMS LASERSAFE™ SPECIFICATIONS

	Continuous wave	Pulsed
Sensitivity	1 to 400 ppm-m*	1 to 400 ppm-m*
Distance	100 m using topographical targets; > 500 m using cooperative targets	Up to 1 km using topographical targets; over 5 km using cooperative targets
Accuracy	Typically better than 5%	Typically better than 5%
Response time	Less than 1 s in most cases	Less than 1 s in most cases
Scan area/field of view	16 degrees x 16 degrees, 50 m x 50 m at range of 200 m	16 degrees x 16 degrees, 280 m x 280 m at 1 km; can also be used for line-of-sight, fixed direction
Spatial requirements	110 V/60 Hz, single phase, <20 A	110 V/60 Hz, single phase, <20 A
Size	106 cm x 112 cm x 70 cm (42 in. x 44 in. x 28 in.)	106 cm x 112 cm x 70 cm (42 in. x 44 in. x 28 in.)
Weight	Approximately 300 lb	Approximately 300 lb
Alarm options	Set at customer-determined levels; audible, visual, automatic telephone dialing	Set at customer-determined levels; audible, visual, automatic telephone dialing
Gases detected	See next page	See next page

*Sensitivity is gas-dependent; 1 ppm-m represents a 1-m gas cloud at 1 ppm or a 10-m gas cloud at 0.1 ppm.

GASES DETECTED WITH ENVIRONMENTAL LASER SYSTEMS LASERSAFE^{TM}

LASERSAFE^{TM} can detect a variety of gases depending on the type of laser used in the system. Most Environmental Laser Systems employ the versatile carbon dioxide laser because of its high efficiency and long life coupled with its reasonable cost and size. The following is a list of gases suitable for monitoring with the carbon dioxide laser:

Acetonitrile
Acrolein
Ammonia
Benzene
Butadiene
t-Butanol
Butene
Chloroethylene
Chloroprene
Cyclohexane
1,2-Dichloroethane
Ethyl acrylate
Ethyl mercaptan
Ethylene
Ethylene oxide
Freon 11,12,113
Furan
Iodopropane
Iso-propanol
Methanol
Methyl chloroform
Methyl ethyl ketone
Methyl mercaptan
Monochloroethane
Ozone
Perchloroethylene
Phosgene
Propane
i-Propanol
Propylene
Styrene
Tetrachloroethylene
Trichloroethylene
Vinyl chloride
Water

This list of gases is not exhaustive and only represents the types of gases monitored.

RECON EXPLORATIONS GASPILS SPECIFICATIONS

Functional

- Sensitivity--methane = <20 ppm-m (7.8 μm) at temperature difference = 4 °C
- Detectable trace gases; methane, ethylene, ethane, nitrous oxide, sulfur dioxide, sulfur hexafluoride, ammonia, carbon monoxide, nitric oxide, nitrogen dioxide, hydrogen sulfide, freons, organophosphates, and any gas with suitable spectral signature in the thermal infrared.
- Interferent rejection (>5,000:1)
- Simultaneous dual gas channel operation as an option
- Closed-circuit television camera and videocassette recorder optional for flight path recovery and data logging

Physical

- Size: head, 36 cm diameter; control, 14 cm H x 25 cm D x 44.5 cm W
- Weight: head, 11 kg; control, 7 kg
- Power: 165 W startup, 28 V dc (100 W operating)
- Environmental: 0 to +40 °C
  flight hardened (vibration, electromegnetic interference, thermal, etc.)
- Head interface compatible with steerable gimbal mounts

Output

- 0 to 10 V dc gas signal for each channel
- 0 to 10 V dc thermal radiance at each channel
- 0 to 10 V dc ratio of dual gas channel signals
- Selectable alarm threshold level
- Analog frequency encoded audio signal suitable for logging into the audio track of a videocassette recorder
- Bar graph display of gas signal
- RS232C interface for displaying decimal values for each gas, radiance and dual gas ratio on a terminal

Options

- Single or dual gas operation
- Closed-circuit television camera and videocassette recorder
- Helicopter or fixed wing aircraft gimbal mount

RECON EXPLORATION PLUMETRACKER SPECIFICATIONS

Principle of measurement	Differential ultraviolet absorption
Detection sensitivity	1 ppm-m, sulfur dioxide and nitrogen dioxide
Calibration stability	±5 ppm-m/24 h, sulfur dioxide and nitrogen dioxide
Dynamic range	1 ppm-m--2,500 ppm-m, sulfur dioxide and nitrogen dioxide
Field of view	1.6 mrad x 60 mrad
Dimensions	20.5 cm x 28.5 cm x 69 cm
Weight	16 kg
Operating temperature	0 to 50 °C
Output	0 to 10 V analog IEEE 488 digital Liquid-crystal display front panel meter

RESEARCH VENTURES, INC., VISIPLUME MODEL 121 SPECIFICATIONS

Range and Accuracy	Range	Accuracy
Sulfur dioxide concentration	0 to 3,000 ppm[a]	± 5 percent FS[b]
Effluent velocity	4 to 70 m/s	± 10 percent FS
Sulfur dioxide emission rate	0 to 4,500 kg/h	± 10 percent FS[b]
Size and Weight	Size	Weight
Optical unit	6 in. od x 24 in. L (9 in. x 9 in. central section)	11 kg
Electronics unit	21 in. L x 17 in. W x 21 in. H	20 kg
Optical unit case (including range correction target)	27 in. L x 18 in. W x 15 in. H	9 kg
Ambient Operating Temperature	0 to 40 °C (32 to 104 °F)	
Power Requirements	12 V dc (120 W); a power supply for 115 V ac (60 Hz) or 230 V ac (50 Hz) operation is also provided	
Optical Unit Components	Ultraviolet vidicon camera, interference filters, neutral density range filters, ultraviolet lens, calibration cell	
Electronic Unit Components	Microprocessor with numeric keyboard, 64-channel digital correlator, cursor positioning controls, television monitor, alphanumeric printer	
Included Accessories	Range correction target, heavy-duty tripod, Polaroid™ camera, viewing hood	
Data Modes	Visualization, calibration, manual, automatic (0.2 and 1 min^{-1})	
Outputs (metric or English)		
Monitor display	Sulfur dioxide concentration, effluent velocity, sulfur dioxide emission rate	
Printer	Identification number, date and time, stack diameter, plume temperature, slant range, sulfur dioxide concentration, effluent velocity, sulfur dioxide emission rate	

(continued)

RESEARCH VENTURES, INC., VISIPLUME MODEL 121 SPECIFICATIONS (continued)

Warranty	Parts and labor for 1 year (excluding vidicon tube warranted by the original manufacturer)
Delivery	90 days after receipt of order

Reseach Ventures, Inc., reserves the right to make changes in design and specifications without prior notification.

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[a] For an assumed path of 2 m.

[b] Typical value for clear day, 200-m slant range, and opacity not exceeding 20 percent.

WRIGHT AND WRIGHT LONG-PATH INFRARED MONITOR MODEL DVX SPECIFICATIONS

Housing Size

Transmitter and receiver--29.5 cm (11.6 in.) height; 39.1 cm (15.4 in.) width; 24.6 cm (9.7 in.) diameter

Housing Weight

Transmitter: 16.8 kg (37 lb); receiver: 18.1 kg (40 lb)

Mounting

3/8-16 in. bolts (4 per housing) to a vertical plate. (Pan-Tilt Mount interfaces between the instrument housings and the user-supplied vertical mounting pipes [4.5 in. outside diameter maximum].)

Operating Voltage

Standard: 105 to 125 V ac, 60 Hz
Optional: 220 to 240 V ac, 50 Hz

Power Consumption

Transmitter: 50 to 250 W (depending on model used)
Receiver: 60 W

Operating Temperature

(Ambient air, with instrument shaded from direct sunlight)

-20 to +43 °C (for wider temperature range, contact factory)

Operating Distance

(Separation between transmitter and receiver)

5 m (16.4 ft) to 100 m (328.1 ft), depending on model used

Volume Monitored

Cylinder with a diameter of approximately 8 cm and length determined by distance between transmitter and receiver

Wright and Wright Specifications (con.)

Sensitivity

Sufficiently sensitive to detect 10 percent lower explosive limit of butane vapors in a 1-m cloud, or 40 ppm over a path length of 50 m, with a signal-to-noise ratio of 10:1 (Sensitivity varies with vapor type; contact factory for additional information.)

Response Time

Operate mode: 63 percent of final reading within 10 to 30 s after a step change in hydrocarbon vapor concentration

Align mode: 63 percent of final reading within 1 to 3 s

Hydrocarbon Vapor Alarm

Single-pole/double-throw relay, rated to 10 A, 120/240 V ac or 28 V dc (Relay changes state when hydrocarbon vapor is within volume monitored and concentration exceeds the factory preset level. Alarm is fail-safe, and setting is field-adjustable.)

Obstruction/Instrument Status Alarm

Single-pole/double-throw relay, rated at 10 A, 120/240 V ac or 28 V dc (Relay changes state when an obstruction blocks the beam or when an instrument malfunction occurs. Relay returns to original state when malfunction is corrected, and alarm is fail-safe.)

Analog Output

Standard: 0 to 10 V dc

Optional: 4 to 20 mA dc

Operation in Hazardous Environments

Housings designed to meet the requirements of Class 1, Division 1, Groups C and D environments; NEMA 4, 7, and 9